U.S. DEPARTMENT OF COMMERCE
National Technical Information Service

PB-272 684

Management of Bottom Sediments Containing Toxic Substances

Corvallis Environmental Research Lab, Oreg

Jul 77

EPA-600/3-77-083
July 1977

PB 272 684

Ecological Research Series

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA. 22161

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.
EPA-600/3-77-083

2.

3. RECIPIENT'S ACCESSION NO.
PB272684

4. TITLE AND SUBTITLE
MANAGEMENT OF BOTTOM SEDIMENTS CONTAINING TOXIC SUBSTANCES: Proceedings of the Second U.S.-Japan Experts Meeting -- October 1976, Tokyo, Japan

5. REPORT DATE
July 1977

6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)
Spencer A. Peterson and Karen K. Randolph, editors

8. PERFORMING ORGANIZATION REPORT NO.

9. PERFORMING ORGANIZATION NAME AND ADDRESS
Environmental Research Laboratory -Corvallis, OR
Office of Research and Development
U.S. Environmental Protection Agency
Corvallis, Oregon 97330

10. PROGRAM ELEMENT NO.
1BA608

11. CONTRACT/GRANT NO.

12. SPONSORING AGENCY NAME AND ADDRESS
same

13. TYPE OF REPORT AND PERIOD COVERED
In House

14. SPONSORING AGENCY CODE
EPA-600/02

15. SUPPLEMENTARY NOTES

16. ABSTRACT

The United States-Japan Ministerial Agreement of May 1974 provided for the exchange of environmental information in several areas of mutual concern. This report is the compilation of papers presented at the Second U.S.-Japan Experts' Meeting on the Management of Bottom Sediments Containing Toxic Substances, one of the 10 identified problem areas.

The first meeting was held in Corvallis, Oregon, in November 1975. The second meeting (at which these papers were given) was hosted by the Japanese Government in October 1976. The 1977 meeting is scheduled for Washington, D.C. in the fall.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
water reclamation sanitary engineering contaminants water pollution ocean bottom sediments freshwater bottom sediments	toxic sediments mercury, PCB contamination of sediments water pollution pollution elimination/control	06/F 08/A,C,J,H 13/B,J

18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS *(This Report)* UNCLASSIFIED	21. NO. OF PAGES 303
	20. SECURITY CLASS *(This page)* UNCLASSIFIED	22. PRICE PC A14 | MF A01

EPA-600/3-77-083
July 1977

MANAGEMENT OF BOTTOM SEDIMENTS CONTAINING TOXIC SUBSTANCES

Proceedings of the Second U.S.-Japan Experts' Meeting
October 1976 -- Tokyo, Japan

edited by

Spencer A. Peterson and Karen K. Randolph
Corvallis Environmental Research Laboratory
Corvallis, Oregon 97330

CORVALLIS ENVIRONMENTAL RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
CORVALLIS, OREGON 97330

DISCLAIMER

This report has been reviewed by the Corvallis Environmental Research Laboratory, U.S. Environmental Protection Agency, and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the U.S. Environmental Protection Agency, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

FOREWORD

Effective regulatory and enforcement actions by the Environmental Protection Agency would be virtually impossible without sound scientific data on pollutants and their impact on environmental stability and human health. Responsibility for building this data base has been assigned to EPA's Office of Research and Development and its 15 major field installations, one of which is the Corvallis Environmental Research Laboratory (CERL), Oregon.

The primary mission of the Corvallis Laboratory is research on the effects of environmental pollutants on terrestrial, freshwater, and marine ecosystems; the behavior, effects and control of pollutants in lake systems; and the development of predictive models on the movement of pollutants in the biosphere.

The United States-Japan Ministerial Agreement of May 1974 provided for the exchange of environmental information in several areas of mutual concern. This report is the compilation of papers presented at the Second U.S.-Japan Experts' Meeting on the Management of Bottom Sediments Containing Toxic Substances, which was hosted by the Japanese Government in October 1976. The first meeting was held in Corvallis, Oregon, in November 1975. The next session is scheduled for Washington, D.C. in the fall of 1977.

A.F. Bartsch, CERL Director
and U.S. Coordinator for the
U.S.-Japan Experts' Meeting

CONTENTS

FOREWORD iii

JAPANESE PAPERS

Dredging of Contaminated Bed Sediment in Japan. 1
T. Sameshima

Countermeasures for Pollution in Tokyo Bay. 20
T. Ohtsuka

An Experiment in Removal of Organically Polluted Bottom Mud from the Seto Inland Sea 62
A. Murakami

The Mechanism of Methylmercury Accumulation in Fish 89
M. Fujiki, R. Hirota and S. Yamaguchi

Determination of Trace Amounts of Methylmercury in Sea Water 96
H. Egawa and S. Tajima

Behavior of Heavy Metals and PCBs in Dredging and Treating of Bottom Deposits 107
K. Murakami and K. Takeishi

A Study on the Behavior of Mercury-Contaminated Sediments in Minamata Bay 127
T. Yoshida and Y. Ikegaki

Using Sand Fill to Cover Dredge Spoils Containing Mercury .144
S. Fujino

Chemical Stabilization of Soft Soils. 155
T. Okumura

A Method for Disposing of Waste Water at Dredged Material Reclamation Sites. 169
E. Satoh

Preceding page blank

UNITED STATES PAPERS

Legal and Administrative Aspects of Bottom Sediment Management 191
A.F. Bartsch

Hydraulic Dredging as a Lake Restoration Technique: Past and Future 202
S.A. Peterson

Interchange of Nutrients and Metals Between Sediments and Water During Dredged Material Disposal in Coastal Waters. . 229
D.J. Baumgartner, D.W. Schults, S.E. Ingle and D.T. Specht

Dredging Conditions Influencing the Uptake of Heavy Metals by Organisms 246
J.F. Sustar and T.H. Wakeman

Dredged Material Densification and Treatment of Contaminated Material 253
C.C. Calhoun, Jr.

Ecological Considerations in Site Assessment for Dredging and Spoiling Activities. 266
D.K. Phelps and A.C. Meyers

APPENDICES

Appendix A--Notes on Units of Measure and Methodology Used by Japanese Authors. 287

Appendix B--Turbidity and Suspended Solids Measurement Methodology 288

DREDGING OF CONTAMINATED BED SEDIMENT IN JAPAN

T. Sameshima*
Technical Counsellor
Bureau of Ports and Harbours
Ministry of Transport

ABSTRACT

Dredging of contaminated bed sediment has only recently started in Japan. In the Sumida River, Tokyo, the first dredging for the purpose of water pollution control was undertaken in 1968. In 1971 a dredging project initiated in Tagonoura port was the first large scale management of accumulated bed sediment in Japan. In 1972-73 the problem of fish contamination was recognized and bed sediments were investigated all over the country. Concurrently, legal and administrative systems concerning pollution control were gradually formulated, and removal of contaminated bed sediments has been extensively undertaken.

This paper discusses the progress of dredging, present status of dredging, and the legal and administrative issues concerning pollution control, especially the cost allocation system which has promoted pollution control efforts.

INTRODUCTION

Since about 1955, when the Japanese economy began to grow rapidly after the post-war confusion, water pollution has gradually become serious. The over-concentration of population and industries in large cities such as Tokyo, Osaka and Nagoya has generated much contaminated sewage which polluted the water and the river beds running through these cities. At that time many rivers were essentially dead. The waters were darkly colored, smelled bad and devoid of fish.

In lakes such as Suwa and Kasumigaura, where the water is neither sufficiently circulated nor exchanged with outside water, eutrophication has been accelerated due to nutrient salts which have flowed into and accumulated in the lakes. Similarly, in the enclosed or semi-enclosed sea areas such as the ports of Tokyo, Osaka, and Nagoya many rivers discharge waste water from nearby factories. This has accelerated the pollution of the water and the bed sediment. At the same time toxic substances contained in effluents from chemical factories and mines accumulated in the sea bed, river bed and agricultural soil and have caused harmful effects on human health through consumption of fish and plants.

---

*Kobe District Bldg., Kaigandori, Ikuta-ku, Kobe 650, Japan

At present, pollution is widely found even in the sea areas around the bays of Tokyo and Osaka and in the Seto Inland Sea. However, restrictions on discharging waste water and sewage and the removal of contaminated bed sediments are considered important efforts toward restoring the former clean environment.

BACKGROUND OF DREDGING

Bed Sediment Contamination

Because of strengthened controls for waste water discharge and the progress of sewage treatment systems the water quality of rivers and ports has been gradually improved and the toxic substances environmental standard for water quality is nearing reality. Some water areas, however, are still considered polluted and cause fish contamination as a result of the accumulated bed sediments which contain toxic substances.

In 1972-73 mercury and PCB contamination of fish was discovered. The intensive accumulation of these substances in fish was caused by the fishes' intake of bed sediment contaminated with these substances. At that time it became an important administrative problem to cope with the pollution of bed sediments. Accordingly, environmental pollution was intensively investigated all over the country in 1972-74 by the Environment Agency, the Fisheries Agency, the Ministry of International Trade and Industry, the Ministry of Transport and the Ministry of Construction in cooperation with concerned local governments. Fish, water, bed sediment and soil were investigated in relation to their pollution by toxic substances such as mercury and PCB.

With regard to mercury contamination, bed sediment was investigated in 332 rivers, 155 ports and 148 sea areas. In total, 5,166 samples were collected and tested from 635 locations. In addition 580 samples from 9 rivers and 3 ports were investigated separately by local governments. The result showed that 278 samples from 19 rivers and 11 ports were contaminated with mercury according to the tentative criteria for removal of bed sediment (7-40 mg/kg) which was established in 1973.

PCB contamination of bed sediment was being investigated throughout Japan in 1972-73. In 1972, out of 2,529 water areas examined, 87 were found contaminated with PCB in excess of 10 mg/kg. In 1973, 1,789 samples were collected and tested from 258 rivers, 38 ports and 58 sea areas. These 354 sample stations showed that 6 water areas were contaminated with PCB over 100 mg/kg, 8 water areas with 50-100 mg/kg, 11 water areas with 25-50 mg/kg and 26 water areas with 10-25 mg/kg (Figure 1, Figure 2).

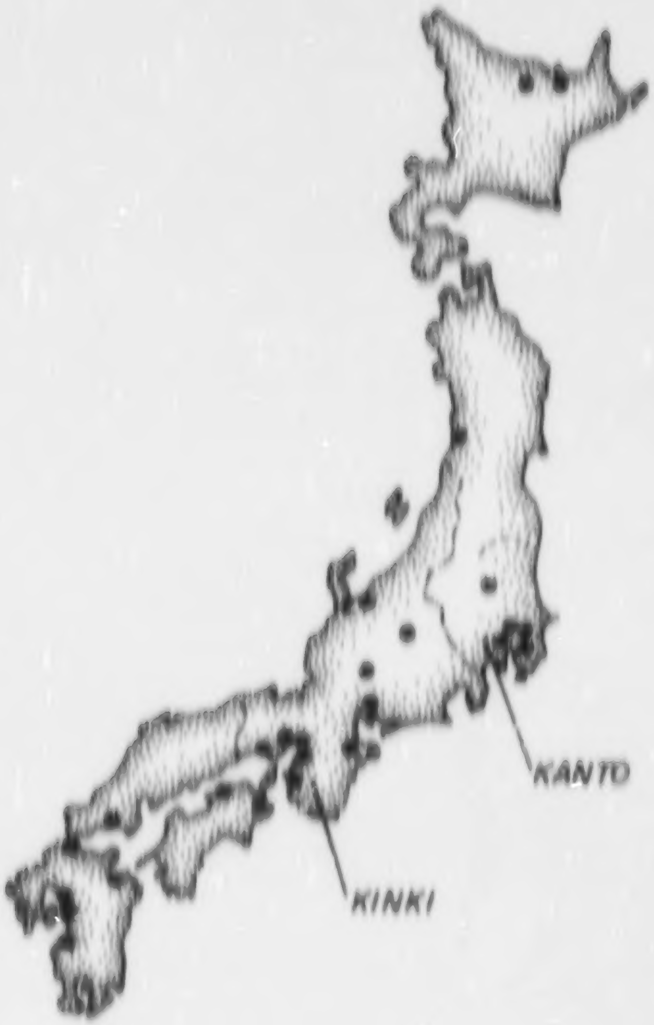


Figure 1. Mercury contaminated bed sediments (1973).

These investigations showed that contaminated bed sediments were distributed all over the country, but contamination with mercury was found to be heaviest in the districts of Kanto and Kinki; greatest contamination with PCB was found in the Tokai and Hokuriku districts.

Bed sediments contaminated with organics were found in rivers running through cities, especially in small, stagnant rivers, and also in canals, creeks and small craft basins.

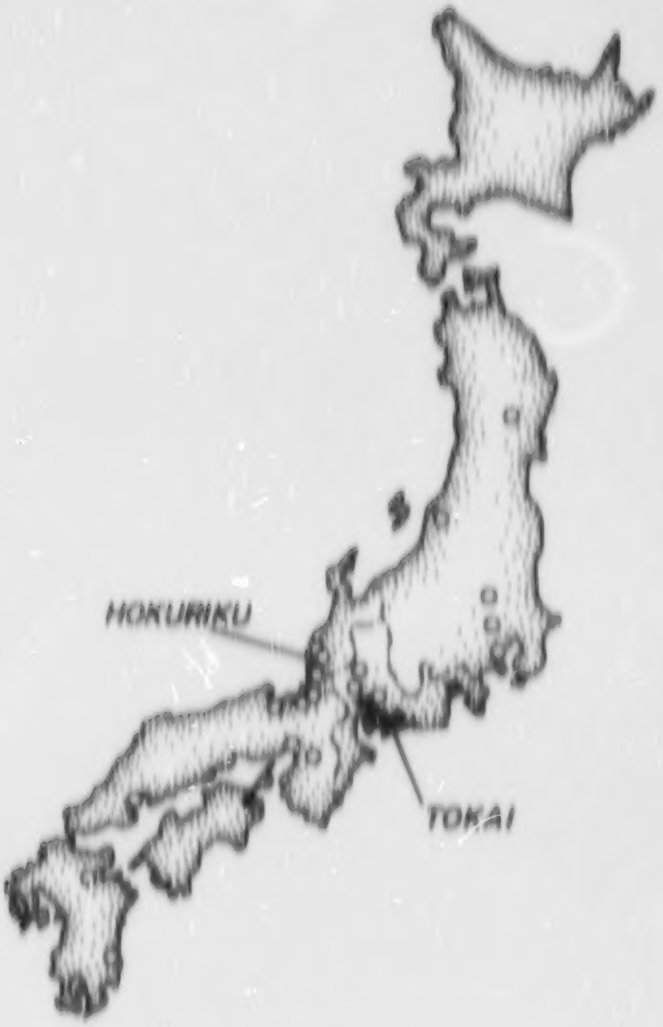


Figure 2. PCB contaminated bed sediments (1973).
● PCB concentration (max) greater than 100 mg/kg
○ PCB concentration (max) 25-100 mg/kg

Progress of Dredging Projects

Contaminated river bed sediments were first dredged in 1958 in the Sumida River. Dredging bed sediment and projects for channeling fresh water followed in rivers which ran through large cities such as Osaka and Fukuoka.

Except for small-scale maintenance dredging in ports located at the mouth of large rivers, dredging of contaminated bed sediment in ports was first undertaken in the port of Tagonoura, Shizuoka prefecture, where the basin was contaminated and silted with pulp mill waste.

The pollution of Tagonoura port became widely known in Japan because the bad odor from the waste water was offensive. So much waste was discharged that it interfered with port activities and damaged the nearby fishery. This event prompted the establishment of the legal and administrative system for anti-pollution in Japan.

At the end of 1970 the "Basic Law for Environmental Pollution Control" was revised and the "Cost Allocation Law for Pollution Control Works," the "Water Pollution Control Law" and the "Marine Pollution Prevention Law" were enacted. Also in 1971 the "Law Governing Government's Special Financial Measures Relative to Pollution Control Projects" was enacted.

In addition to establishing the legal system in 1971 the Environment Agency was created to promote a comprehensive and positive environmental administration which had formerly been under the control of several governmental organizations. At that time dredging of bed sediment containing toxic substances such as mercury became an urgent problem. Therefore, the Environment Agency established "a tentative criteria for removing bed sediment contaminated with mercury," taking into consideration the solubility of mercury from bed sediments agitated by waves and currents, the concentration of accumulated mercury in fish and shellfish, and the eating habits of local inhabitants.

Based on investigation of the PCB concentration in bed sediment and fish, "a tentative criteria for removing bed sediment contaminated with PCB" was established in 1975 to be 10 mg/kg dry weight of bed sediment.

For dredging, it is important to prevent secondary pollution such as diffusion of contaminated bed sediment and leakage of toxic substances from a spillway or containment wall. A "tentative guideline for managing contaminated bed sediment" was established in 1974. This included a study of water quality in selected areas which had already been dredged under the tentative criteria and guidelines.

In addition to PCB and Hg it is also necessary to remove bed sediment contaminated with organic compounds and oil. However, criteria for dredging bed sediments contaminated with organic compounds and oil are not uniformly determined yet, and individual criteria for removing these sediments is left to the implementing organization according to local conditions.

In 1975 the Central and local governments began dredge projects for pollution control in about 70 areas in addition to dredging done by private enterprises at their own cost.

TABLE 1. CHRONOLOGICAL TABLE CONCERNING DREDGING

1956	° "Minamata disease" was officially reported.
1958	° "The Law Concerning Water Quality Conservation of Public Water" and "The Industrial Effluent Control Law" were enacted. Dredging first undertaken in Sumida River.
1960	° Fish with offensive odors were found in Yokkaichi port.
1967	° "The Basic Law for Environmental Pollution Control" was enacted.
1970	° Bed sediment in Tagonoura port was brought to public attention.
	° First "Environmental Pollution Control Program" was established in three areas.
	° "Environmental Quality Standard Concerning Water Pollution" was established.
	° "The Cost Allocation Law for Pollution Control Works," "The Water Pollution Control Law" and "The Marine Pollution Prevention Law" were enacted.
1971	° "The Law Governing Government's Special Financial Measures Relative to Pollution Control Projects" was enacted.
	° "The Environment Agency" was established.
	° Red tide increased rapidly in Seto Inland Sea.
	° First large scale dredging was undertaken in Tagonoura port.
1972	° Dredging undertaken in Kitakyushu port.
1972-73	° Mercury and PCB contamination of fish was publicized.
1973	° "A Tentative Criteria for Removing Bed Sediment Contaminated with Mercury" was established.
1972-74	° Bed sediment was investigated all over the country.
1974	° "A Tentative Guideline for Managing Contaminated Bed Sediment" was established.
1975	° "A Tentative Criteria for Removing Bed Sediment Contaminated with PCB" was established.

Investigation and Research on Pollution Control

To promote the development of science and technology which will help prevent environmental pollution, the Central government takes measures necessary to establish an efficient survey and research program. Both Central and local governments are studying pollution prevention techniques, the influence of pollution on human beings and the environment, and monitoring and surveillance techniques. Research is also undertaken by universities and private firms. At present, pollution control problems are being studied by 53 national investigation and research institutes attached to 11 governmental organizations. The principal national institutes are shown in Table 2 with their main research subjects.

TABLE 2. PRINCIPAL NATIONAL INSTITUTES AND THEIR MAIN STUDIES CONCERNING POLLUTION CONTROL

- Institute of Public Health
  - (Ministry of Health and Welfare)
    - Influence on human health by air pollution
    - Behavior of sediments containing heavy metals
    - Mechanism of photochemical reactions
- Industrial Health Institute
  - (Ministry of Labour)
    - PCB concentration and accumulation in human bodies
- Regional Fisheries Research Laboratory
  - (Ministry of Agriculture and Forestry)
    - Influence on fishery products by thermal effluent
- National Institute of Agricultural Science
  - (MAF)
    - Influence on farm products and fish by environmental pollution
- Regional Institute of Industrial Technology
  - (Ministry of International Trade and Industry)
    - Environmental protection in the Seto Inland Sea
    - Monitoring technique for water pollution in coastal areas
    - Treatment techniques on waste water containing heavy metals
- Geological Survey Institute
  - (MITI)
    - Treatment techniques on mine drainage
- National Research Institute for Pollution and Resources
  - (MITI)
    - Dispersion mechanism of smoke
    - Surveillance of water pollution
    - Treatment technique for automobile exhaust gas
    - Treatment and utilization of sludge
- Port and Harbour Research Institute
  - (Ministry of Transport)
    - Treatment technique for contaminated bed sediment
    - Pollutant diffusion due to tidal current.
- Traffic Safety and Nuisance Research Institute
  - (MOT)
  - Purification of automobile exhausts
- Meteorological Research Institute
  - (MOT)
    - Methods of measuring air pollution
    - Environmental impact assessment concerning air pollution
- Public Works Research Institute
  - (Ministry of Construction)
    - Treatment technique of contaminated bed sediment
    - Nitrogen removal technique at sewage treatment facility

These national institutes are studying various pollution problems arising from public activities under their jurisdiction, while the Environmental Agency comprehensively coordinates these studies to promote them efficiently.

In 1974, the National Institute for Environmental Pollution Research was established as a central institute of investigation and research on pollution control in Japan. This institute is attached to the Environment Agency. In this institute, influence on human health by environmental pollution, the process of environmental pollution and monitoring, and surveillance techniques for environmental pollution are being studied. Investigation and research on the management of contaminated sediments are being studied by the Fisheries Agency, the Ministry of Transport and Ministry of Construction.

The Division of Investigation and Development, the Fisheries Agency, and the Ministry of Agriculture and Forestry are studying techniques for preventing the red tide which often occurs in coastal areas. Red tide is most likely due to eutrophication and is not only harmful to commercial marine products but is also destructive to the remainder of the ecosytem. With a view to preventing fishery damage, a withdrawl technique on the red tide and a removal of the bed sediment which is assumed to cause the red tide are being studied in the field.

Port and Harbour Research Institute, Ministry of Transport, is studying dredging and transportation of contaminated bed sediment, especially turbidity due to dredging and dumping of dredged materials. It is also developing dredge and sediment treatment equipment.

Public Works Research Institute, Ministry of Construction is studying properties of bed sediment and the management techniques for bed sediment, especially bed sediment contaminated with heavy metals and PCB.

In addition to these governmental organizations, many private firms are studying management techniques for contaminated bed sediment including development of dredges, treatment of muddy water and spill water, and prevention of leakage at diked spoils areas.

PRESENT STATUS OF DREDGING

Rivers are under the control of the River Bureau, Ministry of Construction. Dredging of contaminated bed sediments in rivers was put into practice by the Central and local governments in 1958 before a legal and administrative program concerning pollution control was established. Both dredging of bed sediment and improved water systems have been undertaken for water purification in rivers adjacent to large cities, for example the Sumida River in Tokyo and the Kanzaki River in Osaka. Good results were obtained and recently these projects have been undertaken all over the country.

The total volume of bed sediment removed from 1958 to 1975 is about 17 million cubic meters and from 1958 to 1968 the average volume of sediments removed was about 500 thousand cubic meters annually. Recently, the volume increased to 600-1,200 thousand cubic meters annually. In 1975 dredging was done in 53 rivers and lakes such as the Tama River, the Kanzaki River and the Suwa Lake, and a sediment volume of about 700 thousand cubic meters was removed at the expense of 1,945 million yen.

The cost of dredging is borne by those enterprises responsible for the pollutants to the extent of their responsibilities and the remaining cost is publicly shared. Generally, bed sediments in rivers are polluted by many unspecified activities as well as sewage. Dredging is usually undertaken for both river improvement and pollution control. Therefore most dredging in rivers is executed by the Central or local government.

In a Class "A" river which is controlled by the Central government, excluding certain designated sections, the Central government implements river improvement projects directly, which includes dredging of bed sediments. The cost of such projects is shared equally by the Central and local governments. In the case of designated sections of Class "A" rivers and Class "B" rivers, the local government executes river improvement works; one-third of the cost is subsidized by the Central government and the remaining two-thirds is borne by the local government.

If the dredging is listed in the environmental pollution control program, or designated by the Minister of Domestic Affairs as projects to prevent or control pollution, the cost of such projects is shared equally by the Central and local government. For dredging in rivers, shovel type dredges are used in addition to suction and grab type dredges, depending upon the volume and quality of the sediment and surrounding local conditions. Dredge spoils are usually disposed of in containment areas in the sea and, in some cases, in a dry riverbed or on farm land. It is becoming more difficult to find disposal sites, especially inland.

Dredging in Ports

Ports are under the control of Ports and Harbours Bureau, Ministry of Transport.

In Tagonoura port, dredging of contaminated bed sediment was first undertaken by the Shizuoka prefectural government in 1971. Since then dredging of contaminated bed sediments in ports has been executed by port administrations in 16 ports and is already complete in the ports of Mikawa, Sakata, Matsuyama and Aburatsu.

The total volume of bed sediment removed during 1971-75 was 4.5 million cubic meters. In 1976, removal projects were begun in 12 ports such as Tokyo, Osaka and Ohmuta and about 700 thousand cubic meters of sediment were removed in a year at a cost of 8.2 billion yen. The Central government and the port administration bear the costs equally and in each case the enterprise responsible for pollution bears a part of the cost. Both of the government agencies bear the remainder half-and-half. Dredging done by private enterprises at their own cost have already been completed in ports of Chiba (Hg), Kawasaki (Hg), Higashiharima (Hg), Tokuyama-Kudamatsu (Hg), Shimonoseki (Cd), Tsuruga (PCB), Mikawa (PCB), Iwakuni (PCB), and so on.

For dredging in ports, suction type and grab type dredges are widely used. Bed sediments are carried by barge, pipe and truck and are usually disposed into containment areas in the sea. From 1976 to 1980, port improvement works are being carried out along the 5th Five-Year Port Improvement

Plan which was formulated according to the national economy plan. Pollution control works in port will be carried out in parallel with improvement of port facilities based on this plan. These works will be executed in 29 ports including work in progress during this five years (Figure 3).

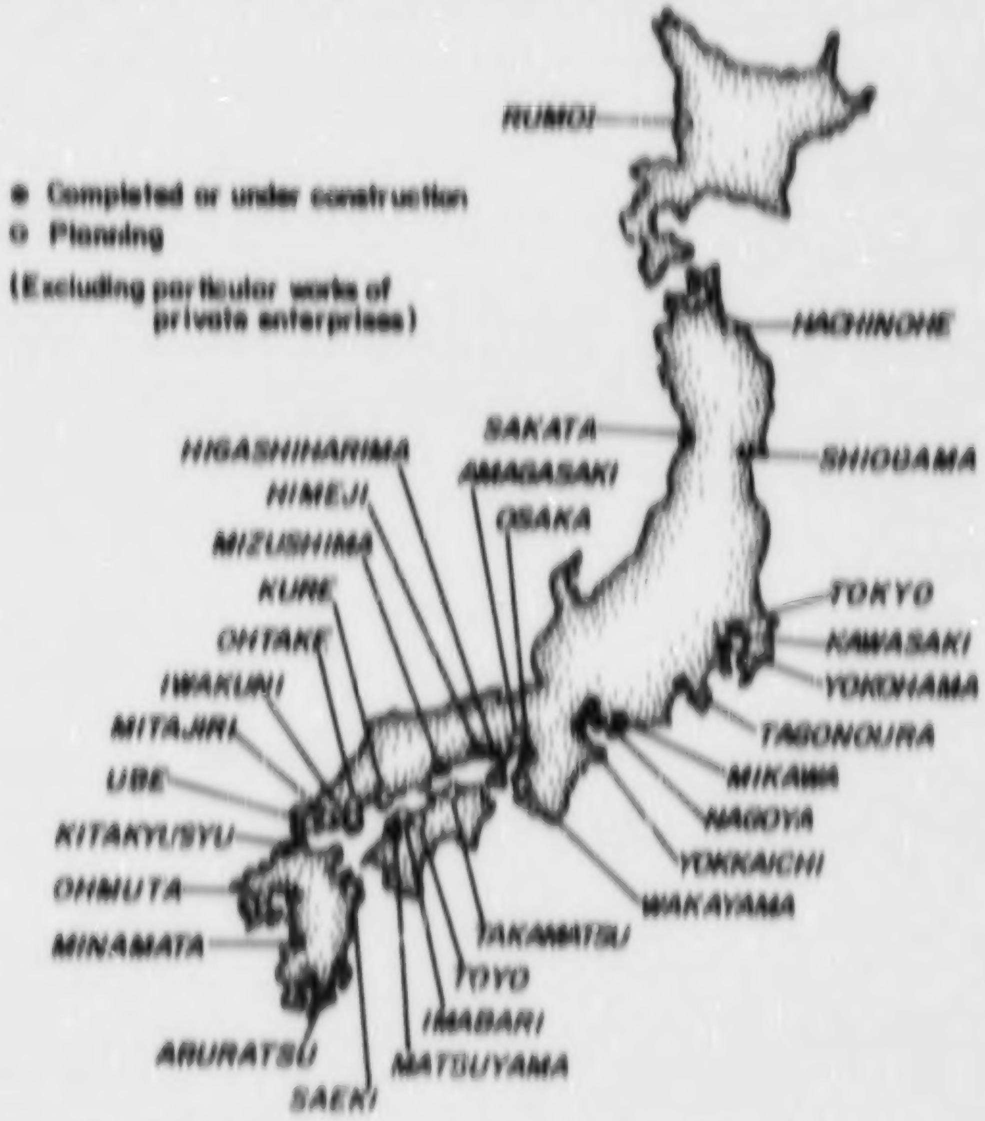

● Completed or under construction
○ Planning
(Excluding particular works of private enterprises)
RUMOI
HACHINOHE
SAKATA
SHIOGAMA
HIGASHIHARIMA
AMAGASAKI
HIMEJI
OSAKA
MIZUSHIMA
KURE
OHTAKE
IWAKUNI
MITAJIRI
UBE
KITAKYUSYU
OHMUTA
MINAMATA
ABURATSU
SAEKI
MATSUYAMA
IMABARI
TOYO
TAKAMATSU
WAKAYAMA
YOKKAICHI
NAGOYA
MIKAWA
TAGONOURA
YOKOHAMA
KAWASAKI
TOKYO

Figure 3. Distribution of ports undertaking removal works (dredging).

In this five-year plan, the total volume of contaminated bed sediment to be removed in 26 ports is estimated at about 12 million cubic meters and the area of contaminated bed sediment to be covered with good soil in Nakatsu and Ohmuta port is estimated at about 500 thousand square meters. Total cost of these works amounts to about 59 billion yen. Of the total cost, 14 billion yen (23.7%) will be borne by the Central government, 14 billion yen (23.7%) by port administration and 31 billion yen (52.6%) by responsible enterprises. In 14 out of 28 ports a part of the cost of these works will be borne by responsible private enterprises. The outline of dredging projects in ports is shown in Table 3.

Dredging in Other Water Areas

There are a few examples of dredging works in water areas other than rivers, lakes and ports. In fishing ports which are under control of the Fisheries Agency, local governments execute dredging projects as pollution control projects with subsidy from the Central government. For example, in 1975-76 a bathing beach was created from a dredging project using contaminated bed spoils covered with good sand.

LEGAL AND ADMINISTRATIVE PROGRAM FOR POLLUTION CONTROL PROJECTS

Both regulation of effluent discharges, smoke, soot and exhaust fumes and execution of pollution control projects such as dredging contaminated bed sediment are required to prevent environmental pollution and to preserve a good environment. Regulation of effluent is prescribed by the Water Pollution Control Law and regulation of smoke, soot and exhaust gas is prescribed by the Air Pollution Control Law. Matters concerning pollution control works are given in the Basic Law for Environmental Pollution Control and the Cost Allocation Law for Pollution Control Works. The latter two laws, with examples of cost allocations, are presented in this chapter.

The Basic Law for Environmental Pollution Control

The Basic Law for Environmental Pollution Control was enacted to identify the responsibilities of private enterprise, the Central government, and local government with regard to environmental pollution control, and to determine the fundamental requirements for control measures to promote comprehensive policies for coping with environmental pollution, thereby ensuring the protection of public health and conservation of the living environment.

This law mainly defines fundamental policies for environmental pollution control. Specific applications for particular control are left to individual laws such as the Water Pollution Control Law, the Air Pollution Control Law, etc. except for the establishment of Environmental Quality Standards and formulation of Environmental Pollution Control Programs in accordance with the Basic Law.

Environmental Quality Standards

At present Environmental Quality Standards, which are maintained for the protection of human health and for the conservation of the living environment, are established with regard to six environmental factors: air pollution, carbon monoxide, sulfur dioxide, noise, aircraft noise and water pollution.

TABLE 3. PLAN OF REMOVAL PROJECTS IN PORTS

Port	Working Period	Removal Volume m^3	Polluting Substances	Main Polluter	Dredge Type	Transportation Method
Tokyo	1978-79	2,402,000	organic compound & sulfide	unspecified	grab	barge
Kawasaki	1980-81	300,000	ditto	unspecified	grab	barge
Yokohama	1973-78	690,000	organic compound & sulfide	unspecified	grab	barge
Nagoya	1972-81	738,670	organic compound, sulfide & mercury	unspecified & chemical ind.	grab & suction	barge & pipe
Yokkaichi	1974-76	2,200,000	oil & mercury	oil refinery & chemical ind.	suction	pipe
Wakayama	1976-77	100,000	organic compound	unspecified	grab	barge
Osaka	1973-81	1,645,000	organic compound	unspecified	suction	barge & pipe
Himeji	1974-80	460,000	organic compound	sewage & leather ind.	suction	barge & dump truck
Kitakyushu	1972-81	3,300,000	organic compound & mercury	steel & chemical ind.	grab	barge
Hachinohe	1979	113,000	organic compound	sewage	grab	barge
Shiogama	1972-77	47,000	organic compound	aquatic food ind.	grab	barge
Tagonoura	1972-77	1,720,000	organic compound & PCB	pulp ind.	grab & suction	dump truck & pipe
Amagasaki	1977-81	200,000	organic compound	sewage	suction	barge & dump truck

TABLE 3. (CON'T.)

Port	Working Period	Removal Volume m^3	Polluting Substances	Main Polluter	Dredge Type	Transportation Method
Higashiharima	1978-79	50,000	organic compound	sewage	suction	barge & dump truck
Mizushima	1972-79	813,000	oil	oil refinery, chemical & steel ind.	suction	barge & pipe
Kure	1976-79	200,000	organic compound & sulfide	pulp ind.	suction	barge & pipe
Tadamatsu	1976-79	397,000	organic compound & PCB	pulp ind. & sewage	suction	barge & pipe
Toyo	1977	20,000	organic compound & sulfide	sewage	grab	barge
Imabari	1978-79	30,000	organic compound	sewage	grab	barge
Iwakuni	1979-80	372,000	organic compound	pulp & oil ind.	grab or suction	barge & pipe
Mitajiri	1976-78	444,500	organic compound	pulp & textile ind.	suction	pipe
Ube	1976-79	492,000	organic compound	pulp & chemical ind.	suction or grab	pipe or barge
Saeki	1977-81	805,000	organic compound	pulp ind.	suction	pipe
Minamata	1974-83	1,675,000	mercury	chemical ind.	suction	pipe
Ohtake	1978-81	890,000	organic compound	pulp & chemical ind.	suction	pipe
Kumoi	1979-80	15,000	organic compound	sewage	dragline	—

For example, the Environmental Quality Standard concerning water pollution (which is an environmental quality standard relating to the health of human beings) prescribes the concentration of toxic metals such as cyanides, alkyl mercury, organic phosphorus, cadmium, lead, hexavalent chromium, arsenic, total mercury and polychlorinated biphenyls. This standard should soon be achieved and maintained uniformly throughout the rivers, lakes and seas.

The environmental standard prescribes water quality in terms of hydrogen-ion concentration (pH), chemical oxygen demand (COD), dissolved oxygen (DO), number of coliforms and normal hexane extraction according to 3 types of water utilization in rivers, and 6 types of water utilization in lakes and seas. The types of water areas were classified by the Central or local government according to local conditions of water use. This standard is to be achieved as soon as possible and then is to be maintained.

Environmental Pollution Control Program

The Prime Minister instructs the concerned prefectural governors to formulate a program relating to environmental pollution control measures to be implemented in specific areas where environmental pollution is serious or likely to become serious, and where it is recognized that it will be extremely difficult to achieve effective environmental pollution control unless comprehensive control measures are taken.

Pollution Control Programs were first established in 1970 in three areas where adequate measures against pollution were required. These are Chiba-Ichihara (Chiba pref.), Yokkaichi (Mie pref.) and Mizushima (Okayama pref.). Since then, programs have gradually been established in 38 areas (second to sixth program) and at present the basic policy has been promulgated to 9 more areas (seventh program) by the Prime Minister. The national share of manufacturers' shipments and population in the 41 areas where these programs are already established is 70% and 50%, respectively. These programs are already established in most of the main industrial areas in the country.

Specific measures to be carried out by governments for pollution control, such as a green buffer zone and removal of contaminated bed sediments, are prescribed in these Pollution Control Programs, and the Central and local governments are obligated to take necessary measures for the full implementation of these programs.

Cost Allocation of Pollution Control Projects

The Central and local governments promote necessary projects for the prevention of environmental pollution in addition to private enterprise, which is responsible for taking necessary measures for the prevention of environmental pollution resulting from specific industrial activities. After private industry pays its share for pollution control projects the remainder is paid for at public expense according to the Cost Allocation Law for Pollution Control Works and the Basic Law for Environmental Pollution Control. These projects are performed by the local governments.

Cost Allocation Law for Pollution Control Works

The Cost Allocation Law for Pollution Control Works was enacted pursuant to the Basic Law for Environmental Pollution Control and provides for proper consideration of the scope of pollution control projects, the responsible private enterprise and the extent of costs to be borne by the responsible private enterprises.

Dredging of bed sediment, water conducting projects and/or any other projects undertaken in rivers, lakes, ports or any other public water areas where water is polluted, are defined as pollution control projects. The cost of pollution control projects, in principle, shall be allocated to each enterprise in proportion to the degree and extent of pollution which arises from its individual activities.

The law prescribes that the scope of the enterprise liable to bear the cost of pollution control projects shall be the enterprise which is definitely considered to engage in individual activities which cause or will cause pollution in the area where the said projects take place. According to these rules, the implementer (the Central or the local government) must determine, after hearing the opinion of the Council (which consists of men of learning and experience), a cost allocation plan which includes (1) the kind of pollution control (2) the criteria to be used to determine the enterprises that are to bear the cost (3) the cost of the pollution control projects (4) the total amount to be borne by the responsible enterprise and the basis of computation and (5) any other matters necessary to execute the pollution control project.

In case of any work listed in the Environmental Pollution Control Program or any work designated by the Minister of Domestic Affairs as work for prevention of pollution, the local government is to be given necessary financial subsidy by the Central government according to the Law Governing Government's Special Financial Measures Relative to Pollution Control Projects.

Example of Cost Allocation

According to the Cost Allocation Law for Pollution Control Works, pollution control projects were put into practice as shown in Table 4 from May 1971 (when the law went into effect) to February 1976. This table shows the total cost of pollution control projects at about 68 billion yen and the total cost borne by responsible enterprises is about 38 billion yen. Therefore, the total cost borne by responsible private firms accounts for 56.4% of all costs.

These projects consist of 19 pollution prevention projects for removing bed sediment, 6 pollution prevention projects for managing soil (such as top-soil replacement at a farm) and 7 installations for green buffer zones. The percent of the total cost borne by private enterprises accounts for 71.1% in removal works, 74.4% in top-soil replacement and 28.9% in green buffer zone projects. The reason why the cost-sharing ratio is so low in the case of the green buffer zone is that in cases of sediment removal and top-soil replacement, the enterprises that bear the cost are limited in number and the pollu-

TABLE 4. LIST OF POLLUTION CONTROL WORKS WHERE THE COST ALLOCATION LAW APPLIED
(MAY 1971 - FEB. 1976)

Area	Total Cost Million Yen	Percentage Paid by Private Enterprise*	Number of Enterprises**	Working Period	Polluting Substances
Dredging of bed sediment					
---Ports---					
Tagonoura (Shizuoka) 1st	500	25	158	1971	SS
2nd	1,885	25	154	1971–77	
Kitakyushu (Fukuoka)	1,800	71	19	1972–73	Cd, As, Hg, SS
Shiogama (Miyagi)	420	6.65	8	1972–80	SS
Mizushima (Okayama)	2,800	77.4	21	1973–77	Oil
Ohmuta (Fukuoka)	4,430	77.42	6	1973–75	Cd, Hg, SS
Yokkaichi (Mie)	5,500	83.28	46	1974–77	Oil, Hg, SS, COD
Sakata (Yamagata)	670	76.3	2	1974–75	Hg
Minamata (Kumamoto)	19,336	65	1	1974–83	Hg
---Rivers---					
Ta (Tochigi)	22	32.6	4	1971–73	SS
Tempaku (Mie)	28	43.8	11	1971–80	SS
Tsusen & Kurinoki (Niigata)	400	67	11	1971–74	SS
Hama (Miyazaki)	1,550	75	2	1972–77	Hg
Nakanoi & Yamanoi (Fukuoka)	6	75.44	7	1973	PCB
Hanamune & Yamanoi (Fukuoka)	17	75.44	7	1973	PCB
Ohe (Aichi)	1,302	57.4	2	1973–77	Hg
Chigiri (Ehime)	50	63.2	12	1973–74	SS
Jurada (Osaka)	32	54.9	11	1974	SS
---Water Ways---					
Sada (Osaka)	3	60.8	1	1973	SS
Sakamoto (Osaka)	10	67.4	1	1974	Cu

TABLE 4. CONTINUED

Area	Total Cost Million Yen	Percentage Paid by Private Enterprise*	Number of Enterprises**	Working Period	Polluting Substances
Top-Soil Replacement Projects					
Usui (Gumma)	913	75	1	1972-75	Cd
Ikuno (Hyogo)	428	75	1	1973-74	Cd
Nakano (Nagano)	132	60	1	1973-74	Cd
Kariya (Aichi)	1,500	74.9	2	1973-74	Cd
Aizu (Fukushima)	408	75	1	1974-76	Cd
Tsubokawa (Aomori)	57	75	1	1975-76	Cu
Installation Projects for Green Buffer Zone					
Mizushima (Okayama)	7,290	25	62	1971-77	Air Pollution
Himeji (Hyogo)	6,629	33.3	15	1973-76	Air pollution, noise
Ohe (Aichi)	1,302	57.4	2	1973-77	Air pollution
Tokai (Aichi)	1,447	33.3	undecided	1973-77	Air pollution, noise
Jonan (Hyogo)	436	25	19	1971-73	Air pollution, noise
Sakaide (Kagawa)	4,373	25	8	1974-77	Air pollution
Tokuyama (Yamaguchi)	3,226	33.3	3	1975-78	Air pollution, noise, offensive odor

Remarks: * Percentage of the cost to be borne by private enterprise
** Number of enterprises liable to bear the cost of the works

tion sources are comparatively definite. Green buffer zone pollution sources are usually very broad and the projects will produce additional benefits other than pollution control, for instance, a park.

An example of a cost allocation plan in Yokkaichi port

The port of Yokkaichi is located in the central part of Japan and faces the northern section of Ise Bay. In the uninhabited portion of the port many oil refineries and oil chemical plants have been located since 1963 and at present the Yokkaichi area is one of the largest petrochemical complexes in Japan. Bed sediment in the south of this port has been polluted by industrial effluents containing oil and mercury. Since 1960 fishes were found to be contaminated and have offensive odors. Because of these matters, the Yokkaichi Port Administration decided to remove the contaminated bed sediment under the environmental pollution control program. This work, begun in 1974, will take 4 years and cost 5.5 billion yen. The sediment to be removed is estimated at 2.2 million cubic meters.

In figuring the cost allocation of this work, enterprises which had to bear the cost of the pollution control work were determined by considering the following three conditions, all of which must be simultaneously satisfied. The first one is enterprises which are located in the designated area, the second one is enterprises with effluents of more than 50 cubic meters per day, and the third one is enterprises from which effluents are judged to have influence upon bed sediments which are to be removed from April 1, 1960 to March 31, 1972.

Then the weight of pollution factors in the cost of the project was determined. In considering the purpose of the project, the process of the sediment formation, the influence to the sea and the marine ecosystem, and so forth, these factors were determined to be 40% for mineral oil, 10% for mercury, 25% for SS and 25% for COD. Then each item to be borne by the private sector and public sector was determined as follows:

Private Sector
(a) Mineral oil, total mercury, SS and COD from effluents from the responsible private enterprises

Public Sector
(a) SS and COD from rivers
(b) SS and COD from domestic sewage
(c) Total mercury from agricultural effluent and domestic sewage
(d) Mineral oil, SS and COD from effluent from exempt enterprises
(e) SS and COD from effluent from bankrupt private enterprises
(f) Mineral oil from ships and others

After the above process, each cost to be borne by private enterprises and the public sector was determined as shown in Table 5.

In the case where the responsible private enterprises share part of the cost of a project, a cost allocation plan is fairly determined after careful consideration by the Council, and then the cost allocation plan is officially announced. According to this system, which is based on the so-called "polluter pays" principle, pollution control projects are extensively and fairly implemented. It is expected that the environment is becoming improved in harmony with economic and social development in Japan.

TABLE 5. COST ALLOCATION IN PORT OF YOKKAICHI

	Items	Percentage to be borne	Cost to be borne in thousand yen
Private Enterprises*	Factory Waste Water	83.28	4,580,400
Public Sector	exempt portion from factory waste water**	6.77	
	waste water from exempt enterprises***	0.46	
	waste water from bankrupt enterprises****	0.09	
	domestic sewage	5.17	
	river water	3.12	
	ships and others	1.11	
	Sub Total	16.72	919,600
Total		100.00	5,500,000

Remarks:

* Number of private enterprises to bear the cost is 48.

** The cost to be borne is partly exempted in consideration of the period when the enterprises were not responsible.

*** Small scale enterprises from which effluents are less than 50 cubic meters per day are exempted from bearing part of the cost.

**** Enterprises which have gone bankrupt are exempted from bearing the cost.

As stated above, the cost of pollution control works is usually borne by both responsible enterprise and the public sector and, in some cases, borne by either the responsible enterprise or the public sector, depending upon the cause of the pollution, the responsible period of the enterprise and so forth.

COUNTERMEASURES FOR POLLUTION IN TOKYO BAY

T. Ohtsuka*
2nd District
Port Construction Bureau
Ministry of Transport

ABSTRACT

Tokyo Bay has been instrumental in the development of the Metropolitan area. However, its quality as a resource has been decreasing with the area's rapid industrialization and urbanization. To improve its quality it is essential to improve the Bay environment.

This paper will discuss the impact of pollution on bottom sediment quality, its purifying function, and the effect of improvement projects on the environment.

OUTLINE OF TOKYO BAY

Geography, Topography

The geographic characteristics of Tokyo Bay (the water area to the north of the line connecting Kannonzaki of Yokosuka City and Isone Point of Chiba Prefecture) are shown in Table 1.

TABLE 1. GEOGRAPHIC CHARACTERISTICS OF TOKYO BAY

Length	Approx. 61 km
Width	Approx. 34 km
Maximum Depth	Approx. 70 m at Old Tokyo River of the Uraga Channel and approx. 40 m at the south end of Nakanose.
Average Depth	Approx. 15 m (steep slope from 5 to 10 m, approx. 8% from 10 m to 40 m) sloped from northeast to southwest.
Length of the Coastline	Approx. 170 km
Water Area	Approx. 1,200 km^2 (Futtsu Point)
Shoreline Configuration Index	Approx. 1.61 (the bay becomes closer to a circle as the number is closer to 1) $SCI = \frac{S}{2\sqrt{\pi a}}$ S = measured perimeter of the bay a = calculated circumference of a circle having the same area as the irregular measured perimeter(s).
Volume	Approx. 18.3 km^3
Amount of Fresh Water Flowing into Tokyo Bay	Approx. 9.98 x 10^9 m^3/y

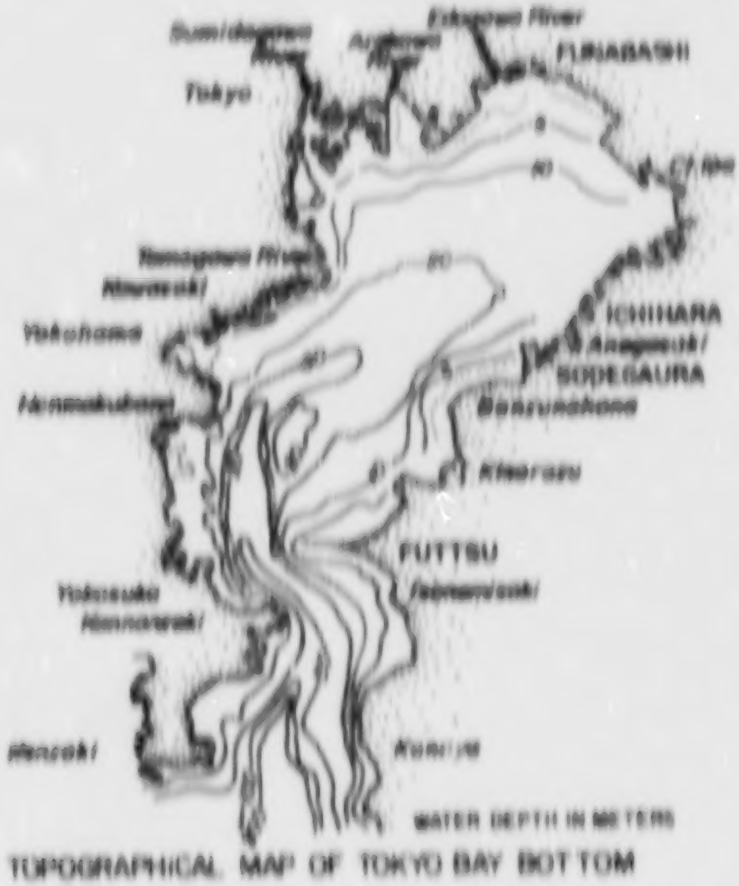
---

*1-2-5 Takashima-cho, Nishi-ku, Yokohama 220, Japan

The length of the Bay is large in comparison to the mouth of the Bay. The water is generally calm and mixing is limited. Characteristic of an enclosed bay, it easily becomes eutrophic.

The coastal area from Yokohama through Tokyo, Chiba and Ichihara has been reclaimed for ports, harbors, industrial sites and housing sites, and thus has lost the characteristic of a natural waterfront. On the other hand, the coastline from Yokohama to Kannonzaki and from Sodegaura to Futtsu excluding Kisarazu, still retains the natural characteristics.

The sea bottom topography shows distinctive characteristics divided by the line connecting Honmoku Point and Futtsu Point. The sea bottom north of the line shows primarily the geographical characteristic of accumulation. The water depth is less than 40 meters and the sea bottom topography is nearly uniform. The sea bottom geology in this area consists of sand in the shallow part and mud in the deep part, with the boundary at the steep slope from 5 to 10 m. In Tokyo Bay, the reclamation is concentrated mostly on the area with a water depth of less than 5 m. The area south of the line has a water depth of about 5 m with exposed bed rock and fewer deposits. The map below shows the topography of Tokyo Bay.


Sumidagawa River
Arakawa River
Edogawa River
FUNABASHI
Tokyo
5
10
Chiba
Tamagawa River
Kawasaki
20
ICHIHARA
Yokohama
Anegasaki/
SODEGAURA
Honmokuhana
Banzuhana
Kisarazu
FUTTSU
Yokosuka
Kannonzaki
Kenzaki
Kanaya
WATER DEPTH IN METERS

TOPOGRAPHICAL MAP OF TOKYO BAY BOTTOM

Present State of Water Quality

About 10 billion cubic meters of water flow into Tokyo Bay from the rivers each year. This means that fresh water equaling approximately half the volume of Tokyo Bay (10.3 billion cubic meters) flows into the Bay each year. The fresh water inflow is becoming more polluted each year; in particular pollution by ammonium nitrogen has been increasing. As a result ammonium nitrogen has been increasing in the Bay. The concentration of COD, BOD and ammonium nitrogen is highest during the winter months of February and March and lowest during July and September. It seems that this is due to the seasonal change in the quantity of river water flow.

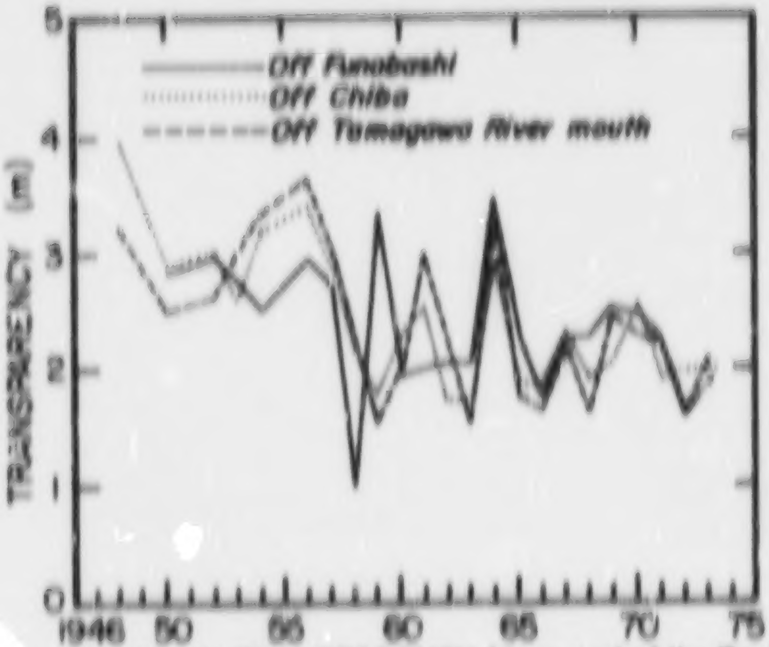
Let us look at the water quality in the sea area. First, the water temperature ranges from 6°C to 28°C during a year. Since clarity is determined by such factors as the quantity of plankton and pollutants, it is used as the most convenient index of water pollution. Figures 1 and 2 show the yearly change of clarity. At the deep end of the Bay, the clarity used to average 3 to 4 meters; it started to decrease from 1958 to 1960. Today it is about 2 meters. Similarly, it was 3.5 to 4.5 meters in the middle of the Bay; today it is about 3 meters. Seasonally it decreases to less than 1 meter in July when red tides appear frequently. Then, it recovers from fall to winter as plankton populations decrease. Clarity is greater at the mouth of the Bay where the ocean sea water has more influence. Since clarity is influenced by the quantity of plankton, it is also an indicator of the stage of eutrophication.

Figure 3 shows that oxygen saturation exceeded 100% almost every year, but there has been a decreasing trend since 1970. The Bay mouth area had a normal percentage of oxygen saturation from 100 to 110 percent, excluding the oversaturation during the development of the red tide in 1968, but it fluctuated widely both in the middle and deep end of the Bay, indicating that eutrophication in the Bay has been progressing.

Next, let us look at organic matter. Figure 4 shows the annual change of COD value after 1963. According to the figure, the value of COD has been within a range of 1 to 3 ppm in general; about 1 ppm at the Bay mouth and 2 ppm in the middle area. Annual fluctuation has been minimal. However, in the deep end of the Bay, it has been increasing slightly since 1966.

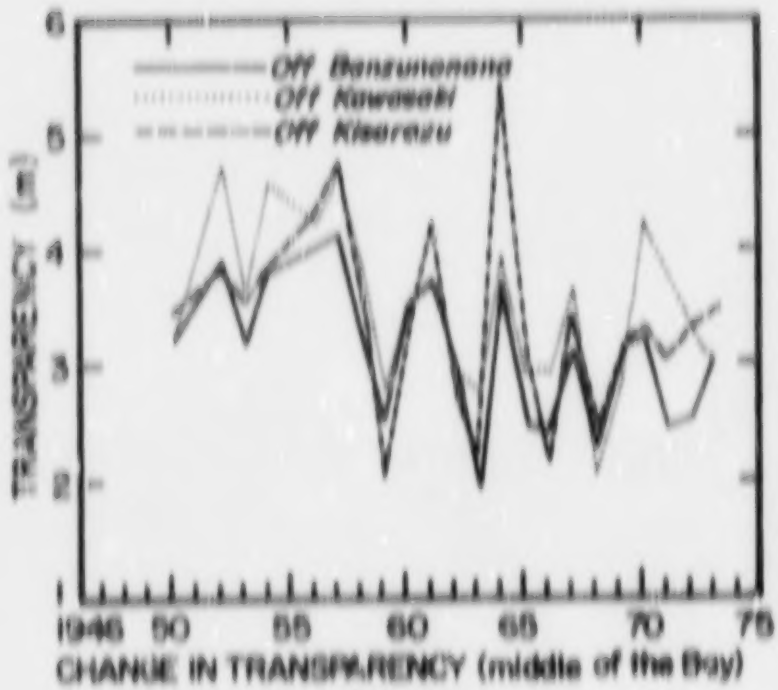
Figures 5 through 7 show the values of nutrient salts such as NH_4-N, NO_3-N and PO_4-P, respectively. From the figures, it is clear that they have been increasing since 1964, especially NO_3-N and PO_4-P.

By area, the highest value of NH_4-N was measured at the deep end of the Bay with around 0.5 ppm, for NO_3-N and PO_4-P, relatively higher values were found in the middle of the Bay with 0.2 ppm, and 0.05 ppm, respectively. While the values of NH_4-N showed considerable variation depending on sea area, neither NO_3-N nor PO_4-p showed such wide variation.



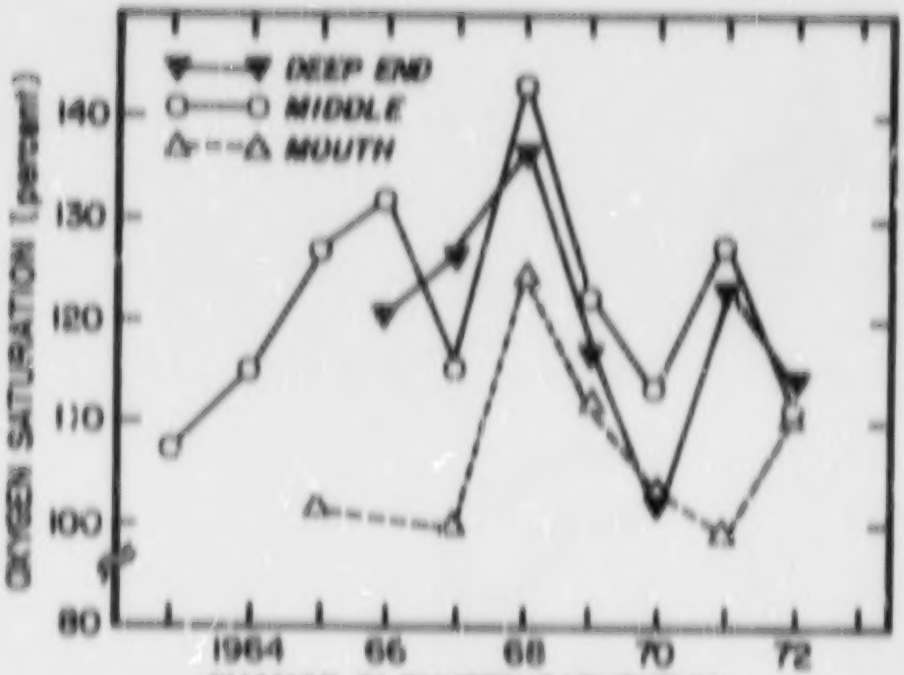
CHANGE IN TRANSPARENCY (deep end of the Bay)

Figure 1



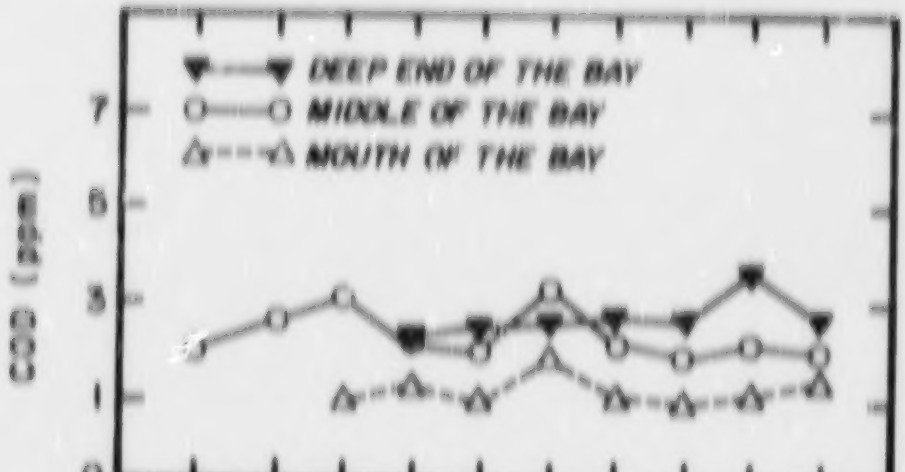
CHANGE IN TRANSPARENCY (middle of the Bay)

Figure 2



CHANGE IN OXYGEN SATURATION

Figure 3



CHANGE IN COD

Figure 4

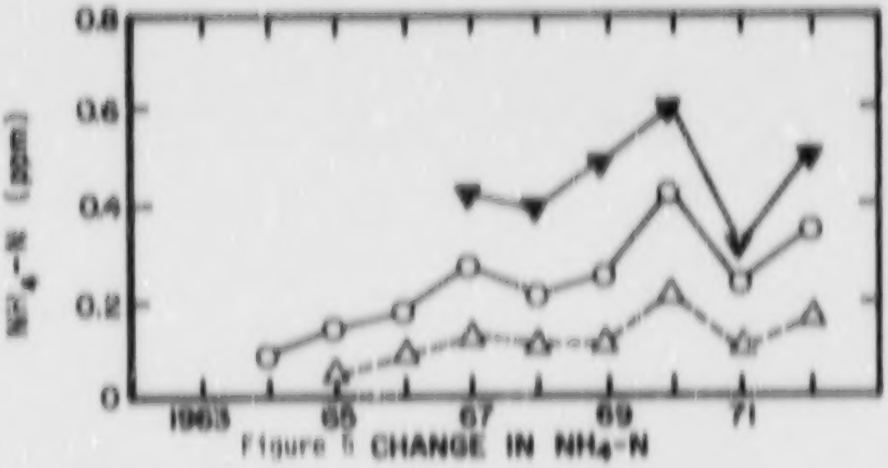


Figure 5 CHANGE IN NH_4-N

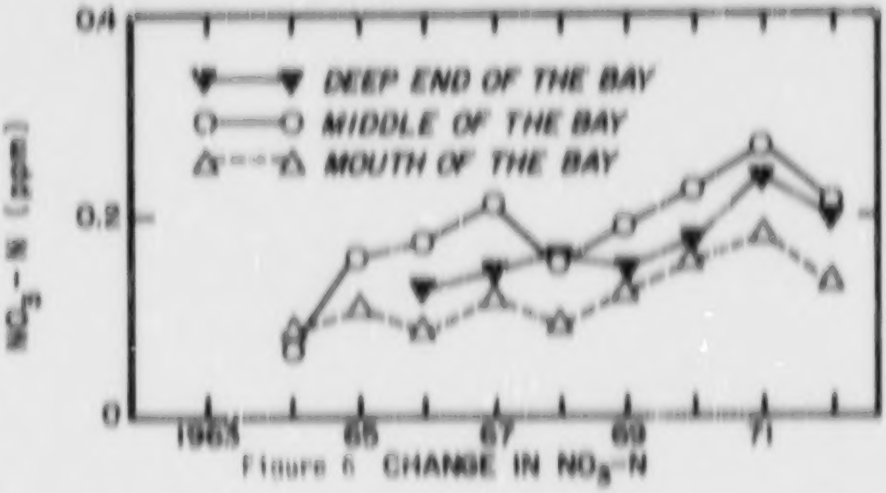


Figure 6 CHANGE IN NO_3-N

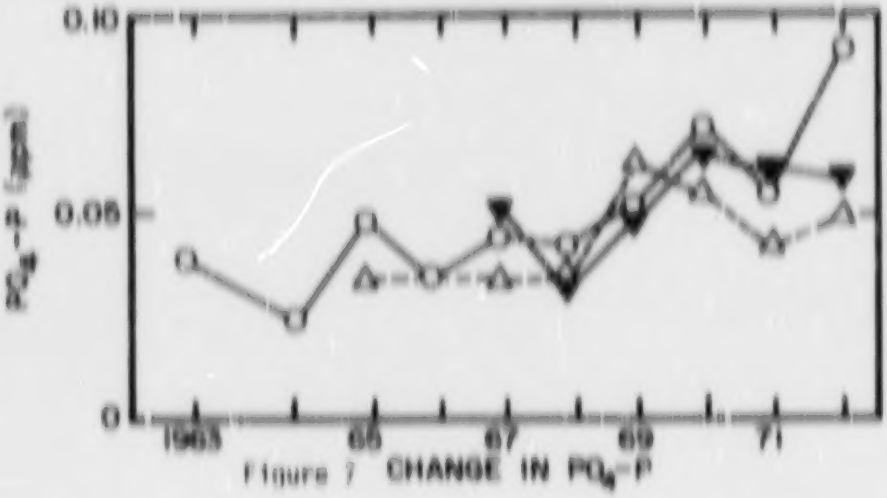
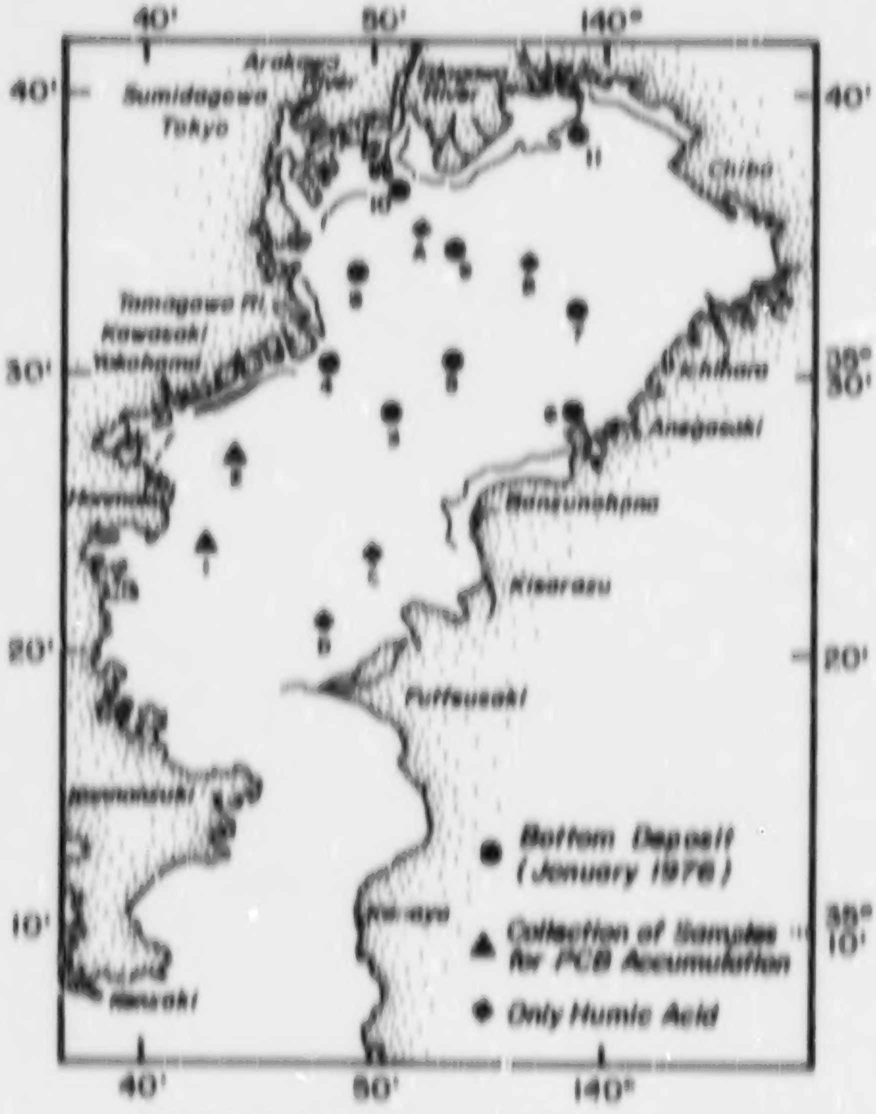


Figure 7 CHANGE IN PO_4-P

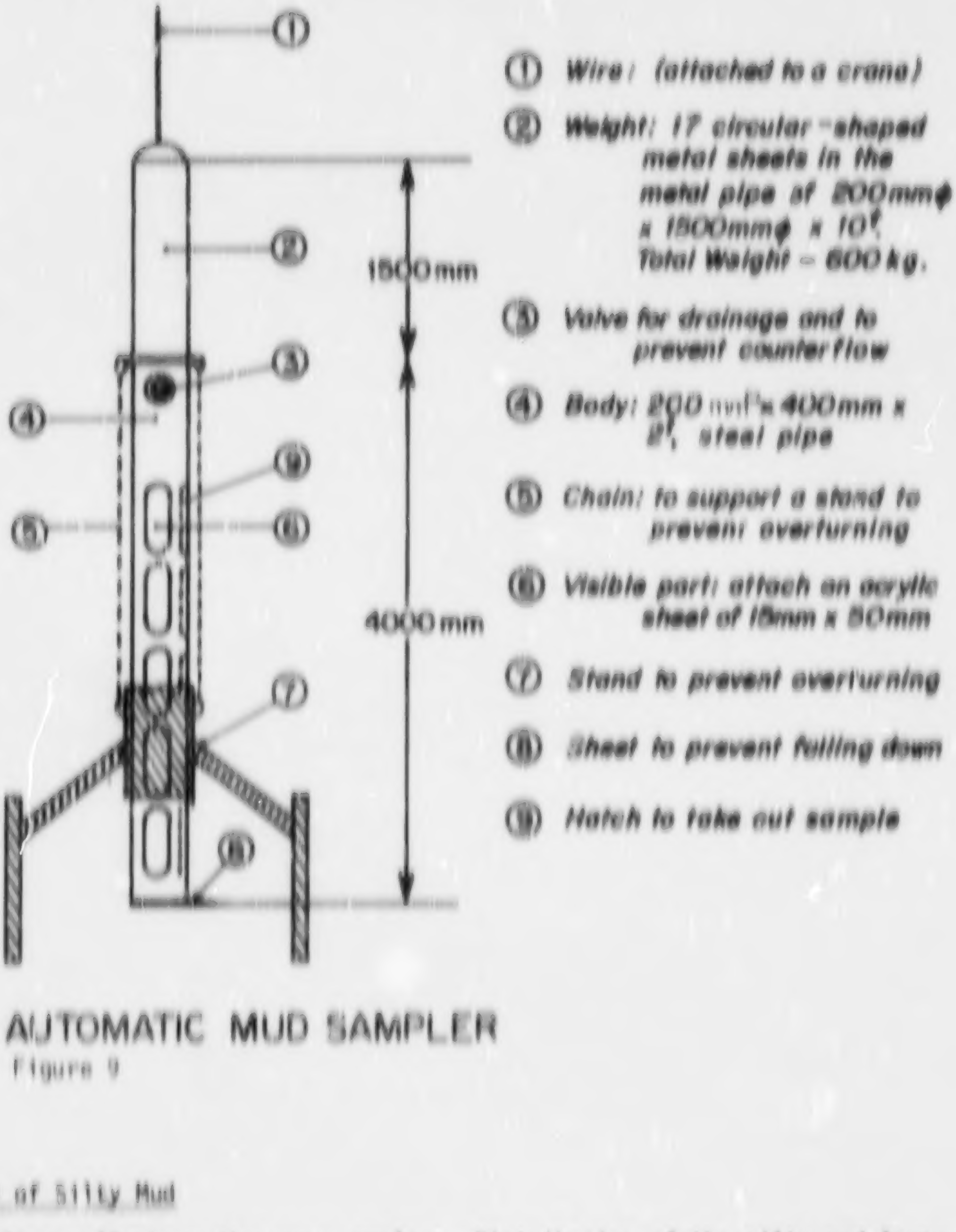
PRESENT CONDITION OF THE BOTTOM DEPOSIT

In January, 1976, a survey was made of the general condition of the bottom deposit in the Bay of Tokyo. The survey was carried out at 11 points (Figure 8) sediments were collected with a mud sampler (Figure 9). Results of the survey are discussed in the following sections.


40'
50'
140°
Arakawa River
Sumidagawa
Tokyo
River
Chiba
Tamagawa R.
Kawasaki
Yokohama
Ichihara
Anegasaki
Kisarazu
Futtsusaki
Kenzaki
35° 30'
20'
35° 10'
Bottom Deposit (January 1976)
Collection of Samples for PCB Accumulation
Only Humic Acid

BOTTOM POINTS SURVEYED

Figure 8


①
②
③
④
⑤
⑥
⑨
⑦
⑧
1500mm
4000mm
① Wire: (attached to a crane)
② Weight: 17 circular-shaped metal sheets in the metal pipe of 200mmφ x 1500mmφ x 10t. Total Weight = 600 kg.
③ Valve for drainage and to prevent counterflow
④ Body: 200 mmφ x 400mm x 2t, steel pipe
⑤ Chain: to support a stand to prevent overturning
⑥ Visible part: attach an acrylic sheet of 15mm x 50mm
⑦ Stand to prevent overturning
⑧ Sheet to prevent falling down
⑨ Hatch to take out sample

AUTOMATIC MUD SAMPLER
Figure 9

Layer of Silty Mud

Figure 10 shows the core samples. Distribution of the silty mud layer is shown in Figure 11, considering the blackened sediment as silty mud. As a result of the survey, it was learned that the silty mud layer was 0.5 m thick, contained 32% mud and the specific gravity was 2.5. Therefore, it was estimated that the quantity of silty mud deposited in the area (approx. 1,000 km²) of Tokyo Bay was about 400 million tons.

CORE SAMPLES

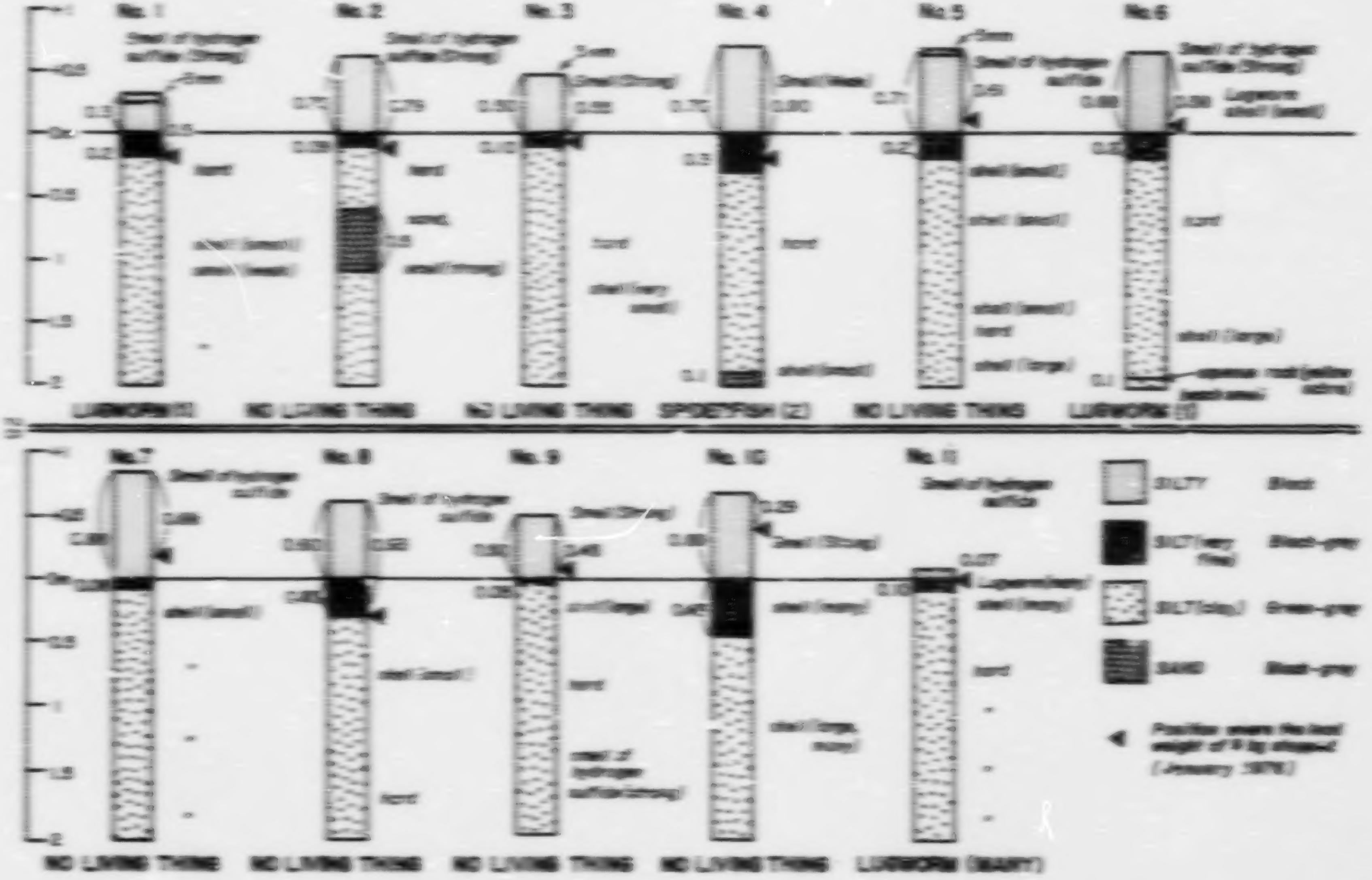


Figure 10

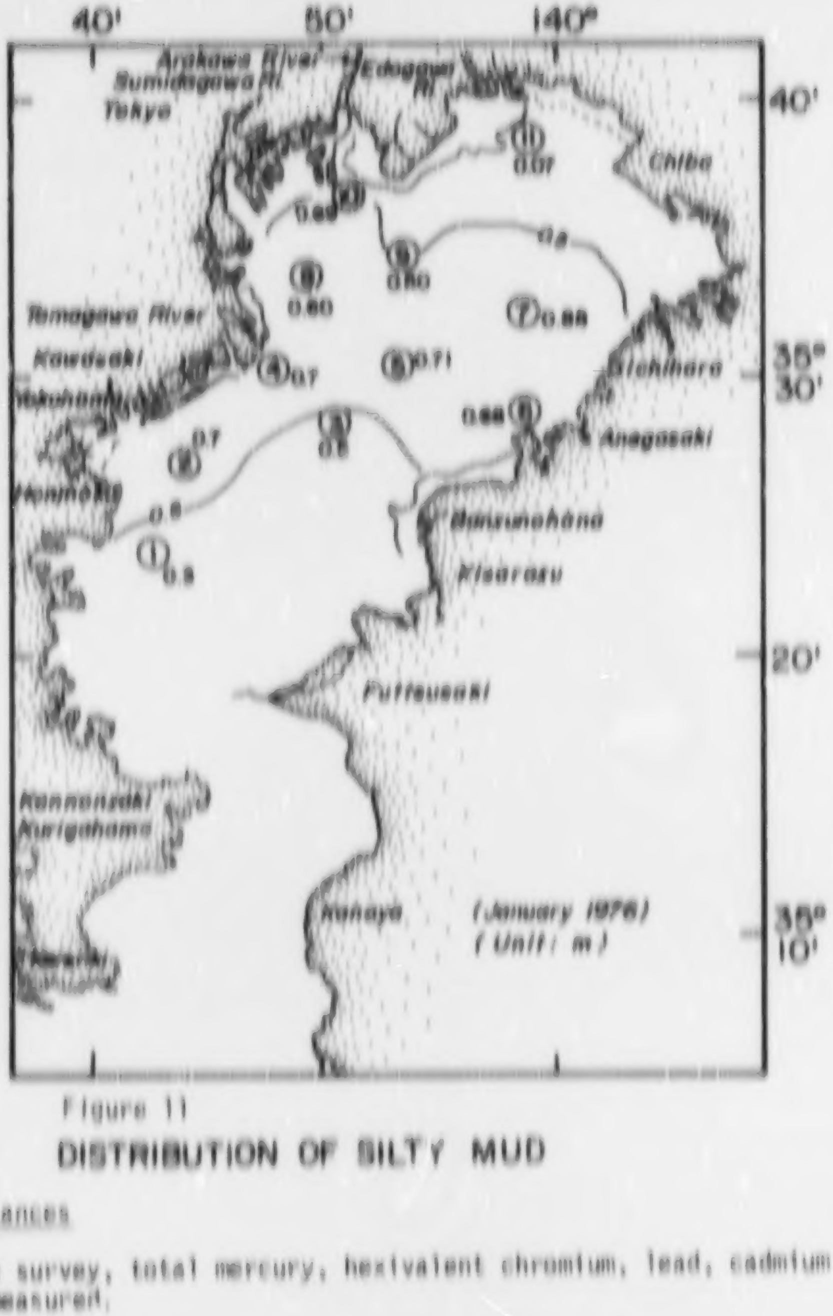

40'
50'
140°
40'
35° 30'
20'
35° 10'
Arakawa River
Sumidagawa Ri.
Tokyo
Edogawa Ri.
Chiba
Tamagawa River
Kawasaki
Yokohama
Honmoku
Ichihara
Anegasaki
Banzunohana
Kisarazu
Futtsusaki
Kannonzaki
Kurihama
Kanaya
(January 1976)
(Unit: m)
0.91
0.80
0.80
0.88
0.7
0.71
0.88
0.7
0.8
0.5

Figure 11

DISTRIBUTION OF SILTY MUD

Toxic Substances

In the survey, total mercury, hexivalent chromium, lead, cadmium and PCBs were measured.

(1) Total Mercury

Figure 12 shows the horizontal distribution of total mercury. The silty mud from the sea areas (No. 4) off Kawasaki and Yokohama (No. 2) contained more than 1.0 mg/kg; high density area extends in a tongue shape from the west side of the Bay towards Aneyaskaki.

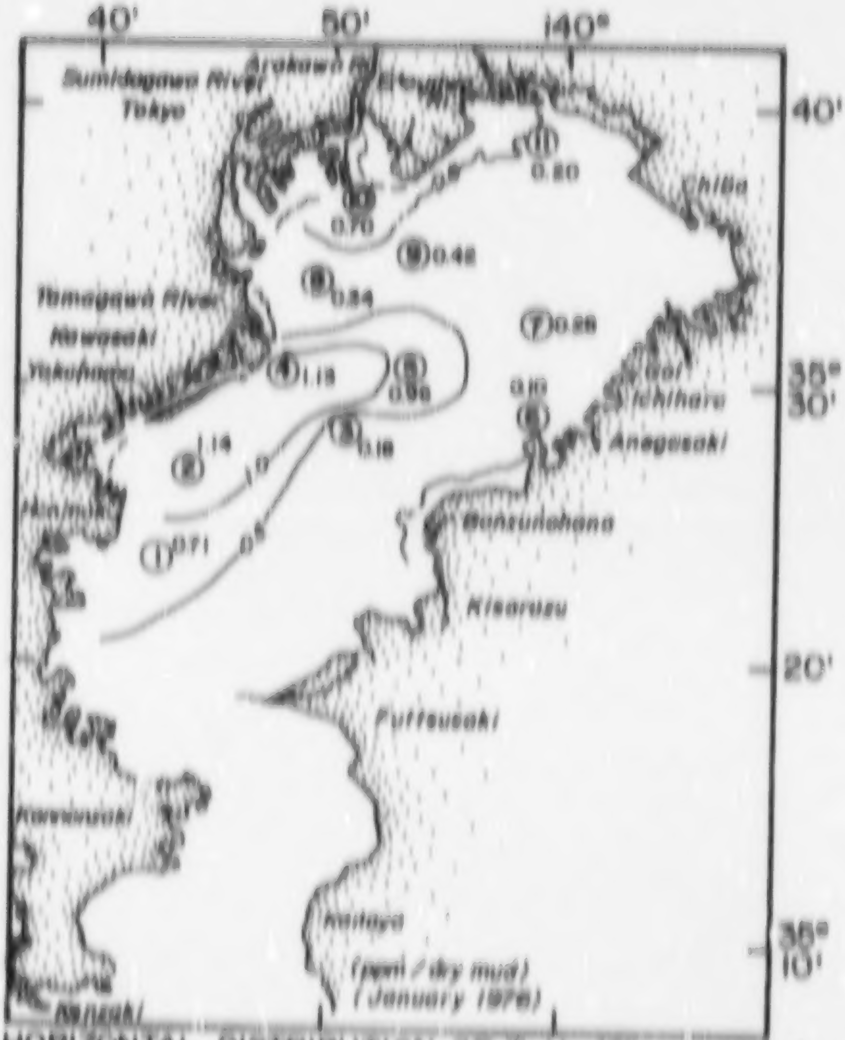

40'
50'
140°
40'
35° 30'
20'
35° 10'
Sumidagawa River
Tokyo
Arakawa R.
Chiba
Tamagawa River
Kawasaki
Yokohama
Goi
Ichihara
Anegasaki
Honmoku
Banzunohana
Kisarazu
Futtsusaki
Kannonzaki
Kaneya
0.20
0.70
0.5
0.42
0.34
0.26
1.15
0.96
0.16
0.18
1.14
1.0
0.71
0.5
(ppm / dry mud)
(January 1976)

Figure 12 HORIZONTAL DISTRIBUTION OF T-Hg (Silty mud level)

Next, Figure 13 shows the vertical distribution at each survey point.

The surface layer which was 50 cm thick showed a high density of mercury, particularly in the silty mud.

(2) Hexivalent Chromium

As far the the result of this survey is concerned the density of hexivalent chromium was less than the quantitative limit of 1 mg/kg at all test sites.

(3) Cadmium

In the silty mud, both at the station (No. 10) off Arakawa River mouth and the stations (No. 4 and No. 5) off Kawasaki, cadmium density was from 2 to 3 mg/kg in the silty mud; at all other points, it was less than 1 mg/kg. Figure 13 shows the vertical distributions.

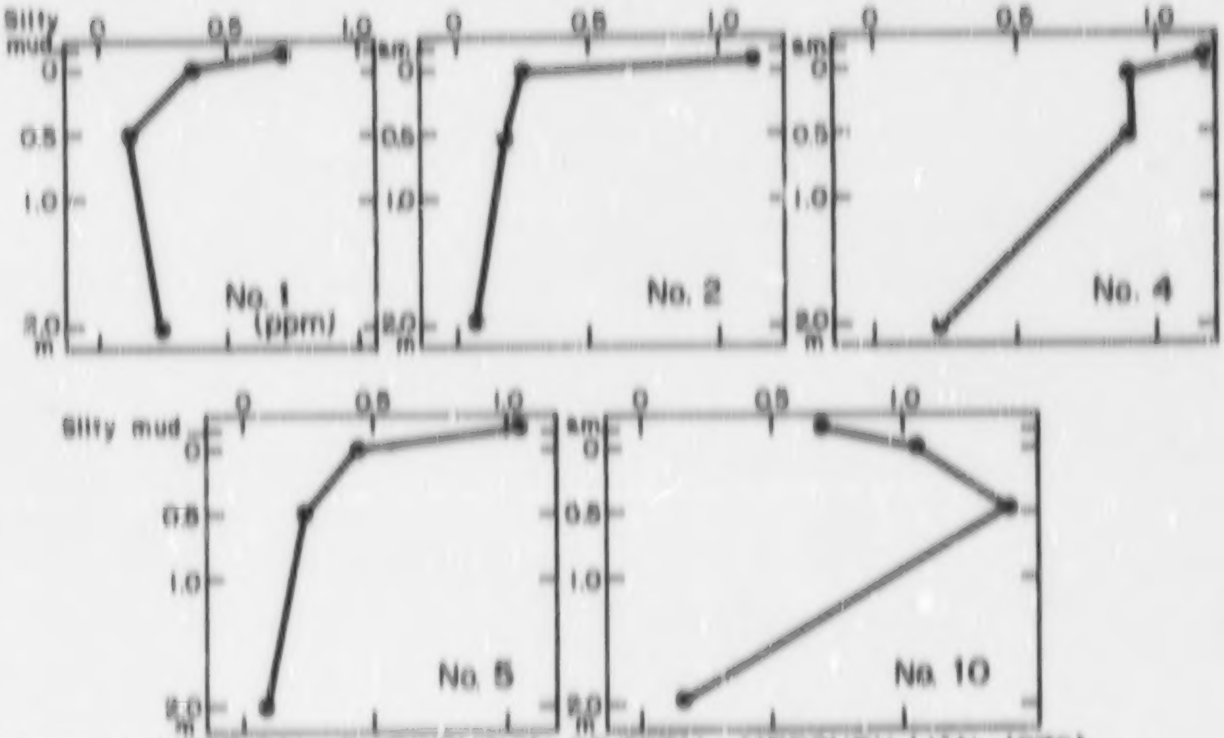


Figure 13 VERTICAL DISTRIBUTION OF TOTAL MERCURY (JAN., 1976)

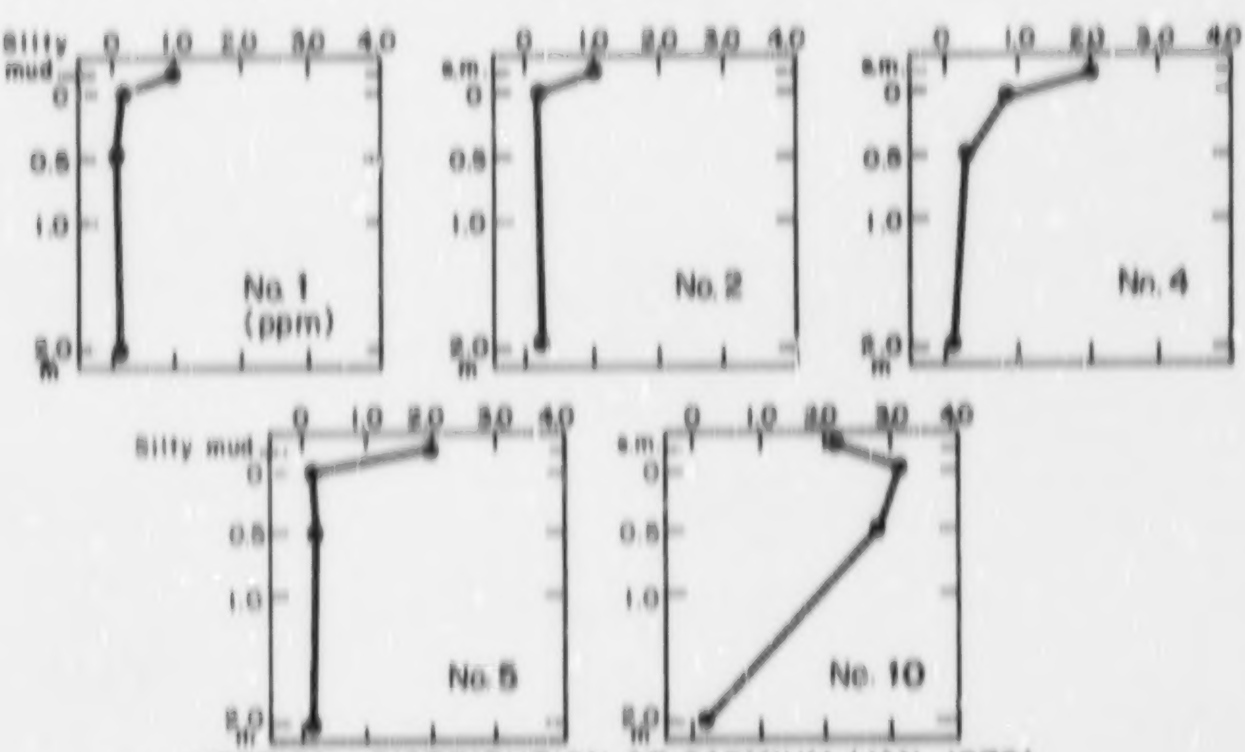


Figure 14 VERTICAL DISTRIBUTION OF CADMIUM (JAN., 1976)

(4) Lead

There was a highly dense area of more than 50 mg/kg of lead from the area off the river mouth of the Arakawa River to the area off the Tamagawa River and Yokohama port. As shown in Figure 15 the vertical distribution of concentration was almost constant at 15 mg/kg in depths greater than 0.5 m, excluding the area off the Arakawa and Tamagawa Rivers. Therefore, this value can be considered as the natural environmental value of lead in Tokyo Bay.

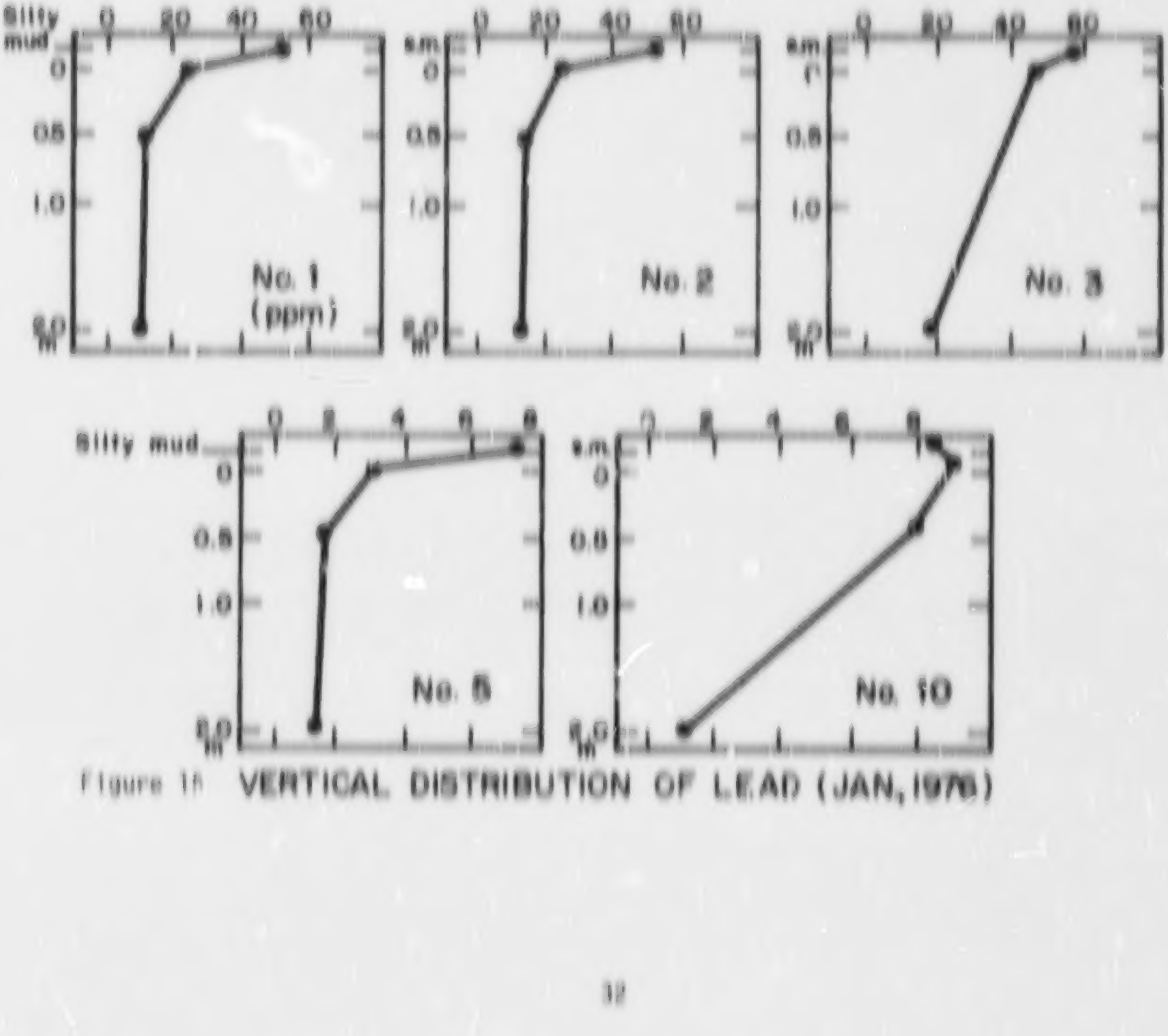


Figure 15 VERTICAL DISTRIBUTION OF LEAD (JAN,1976)

(5) PCB

According to the "Outline of Results of the National Environmental Survey (Water and Bottom Deposits) 1973, PCB", by the Water Quality Bureau of the Environment Agency, the PCB density varied from 0.4 to 28.0 mg/kg with an average of 7.96 mg/kg in the Keihin Yokohama Port. The maximum density of 2.57 mg/kg was detected in the bottom deposits of Tokyo Port. Both of these highly polluted areas were within Tokyo Bay. However, countermeasures to remove PCBs are almost completed and the present values of PCB concentration are from 0.1 to 0.6 mg/kg at Station No. 7 off Yokohama Port and Station No. 1 off Honmoku.

Distribution of total mercury, cadmium, lead and PCBs was discussed above. Relatively high concentrations of these substances were distributed into the bay near the mouths of the Arakawa and the Tamagawa Rivers. From this, it is estimated that toxic substances flow from the rivers into Tokyo Bay in large quantities. Vertically, the concentration was especially high in the silty mud at all points except Station 10 off the Arakawa River mouth. The effect of man-induced pollution was limited to the surface layer of 50 cm.

(6) Oil

According to the Marine Pollution Survey Report (Hydrography Department, Maritime Safety Agency, November, 1974), the condition of oil pollution in the bottom deposits of Tokyo Bay was surveyed with saturated hydrocarbon as an indicator. Oil contains saturated hydrocarbon on the order of several tens of percents. The density of saturated hydrocarbon in the bottom deposit was especially high, both in the northwest sea area and the area off Goi, of Chiba Prefecture. The maximum value was more than 700 mg/kg. The average density was about 300 mg/kg in Tokyo Bay. Judging from these values and the speed of accumulation, it has been estimated that the total quantity of oil (as saturated hydrocarbon), which is artificially discharged, amounts to 1.2 × 10[illegible] m tons. According to the survey, the normal hexane extraction method of analysis found an oil density of 3030 mg/kg in the area off Arakawa River mouth and less than 1000 mg/kg in other areas.

Organic Substances and Nutrient Salts

The results of the survey are shown below.

(1) Percentage of Mud

The percentage of mud contained was 32.4% on the average in the silty mud and 21% in the central points No. 5, 7, 8 and 9. There was a tendency for the percentage to be smaller in the middle than in the coastal region. This tendency was found in other layers, too. Since the areas with low percentages of mud correlated with areas containing thick sludge, it may be inferred that particulates have been settling in the middle of the Bay due to low water velocity.

(2) Ignition Loss

The silty mud showed a rate of more than 10% ignition loss (IL) in all the areas, excluding the deep end coastal region in front of Funabashi Port and off Arakawa River mouth. A maximum of 16.1% IL was observed in the middle of the Bay at Station No. 5. In the area off Haneda, a high rate of 15.6% IL was observed and the general region of high values (over 13% IL) extended from the area off Haneda to the middle of the Bay (Figure 16).

(3) COD

The area with densities over 30 mg/g extended in a circular pattern from the middle of the Bay off Haneda. The horizontal distribution of COD in the silty mud tended to be similar to that of ignition loss and percentage mud, but a distinctive difference was observed in that its vertical distribution rapidly decreased from the silty mud layer to the 0 m level. This was especially noticeable in the middle of the Bay (Figure 17).

(4) Sulfide

An extremely high concentration of 3.73 mg/g was detected at point No. 8 off Haneda and the area with concentrations greater than 1.0 mg/g extended from the west part of Tokyo Bay to Anegasaki. The sulfide distribution was similar to that of the abiotic zone. Vertical distribution showed a drastic decrease from the silty mud layer to the 0 m level and there was no variation in density in the layers below 0 m at all stations except Station 10. At the points off Arakawa River mouth and Tamagawa River mouth, a concentration of over 0.2 mg/g in the 0 m layer was detected, which was considered to have an effect on living things (Figure 18).

(5) Total Organic Carbon

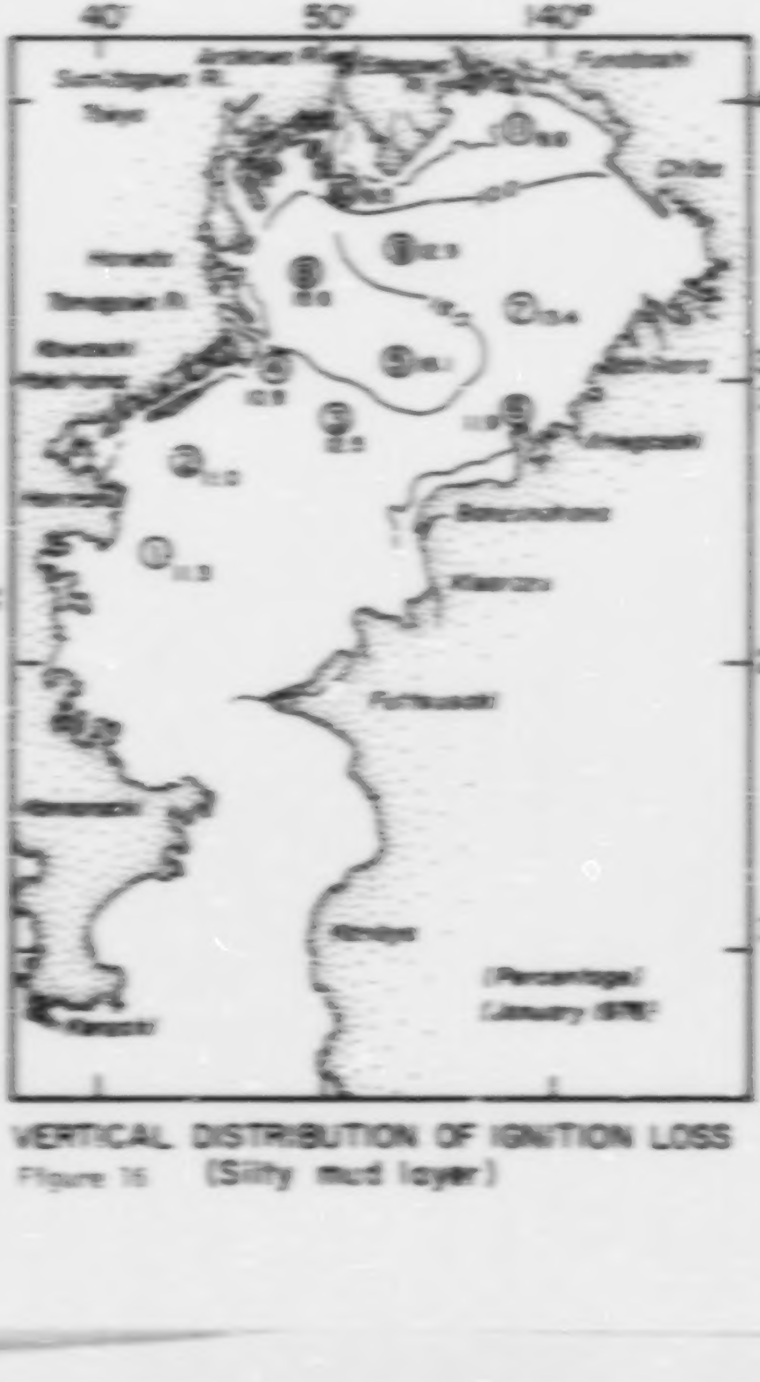
The area extending from Yokohama to Chiba and Tokyo Port showed TOC densities of over 10 mg/g. In the middle of the Bay (Station 5) a rich organic content of 15.2 mg/g was noticed.

(6) Total Nitrogen

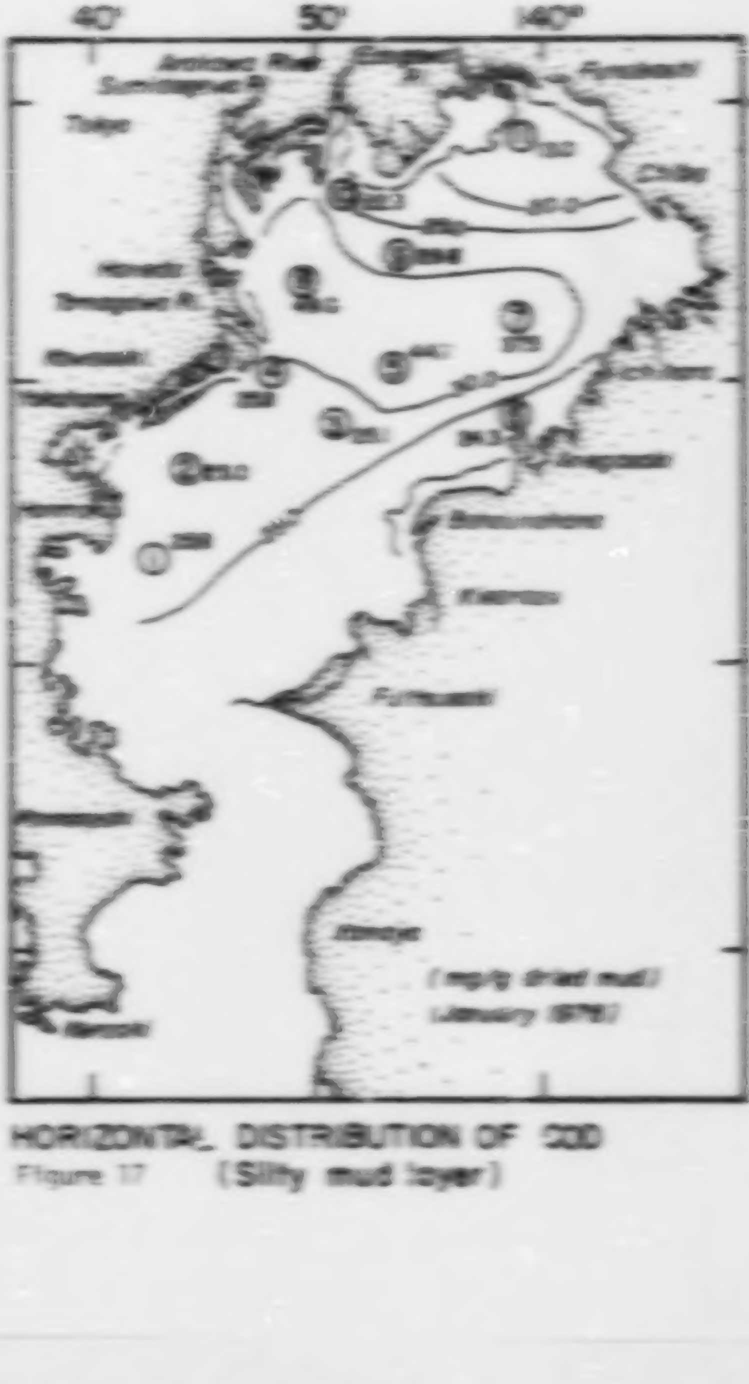
In the silty mud layer, a density of over 2000 mg/kg was observed in the area extending west from Tokyo Bay to Anegasaki. Especially at Station No. 1 off Honmoku the value for N was over 4,000 mg/kg, and the area of high density extended from the middle to the deep end of the Bay (Figure 19). A similar distribution was observed in the 0 m layer.

(7) Total Phosphorous

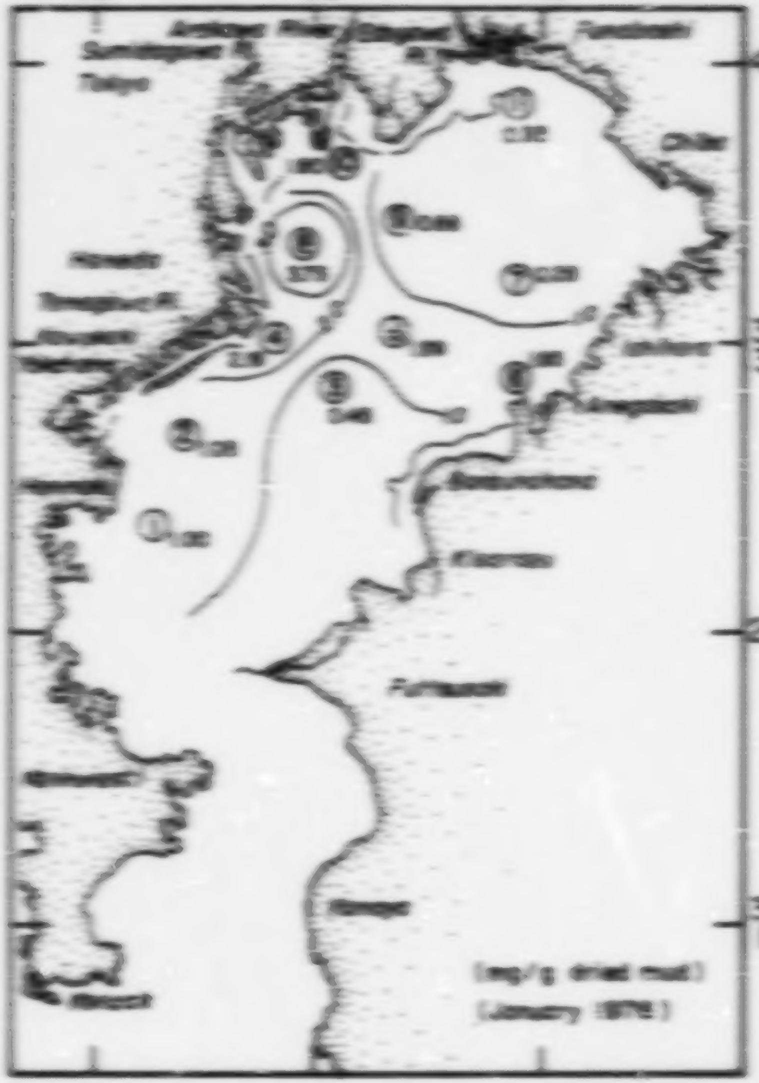
The rivers exert a considerable influence in the case of phosphorus, and therefore a high density is observed at river mouths. In the silty mud layer, a density of over 600 mg/kg was observed in the area extending from Tokyo Port to the mouth of Tamagawa River. The area where values were greater than 500 mg/kg was similar to that of nitrogen. In the 0 m layer, the circular area extending from the mouth of Arakawa River to that of the


40'
50'
140°
40'
35°30'
20'
35°10'
Sumidagawa R.
Tokyo
Funabashi
Chiba
Tamagawa R.
Kawasaki
Anegasaki
Kisarazu
Futtsusaki
(Percentage)
(January 1976)

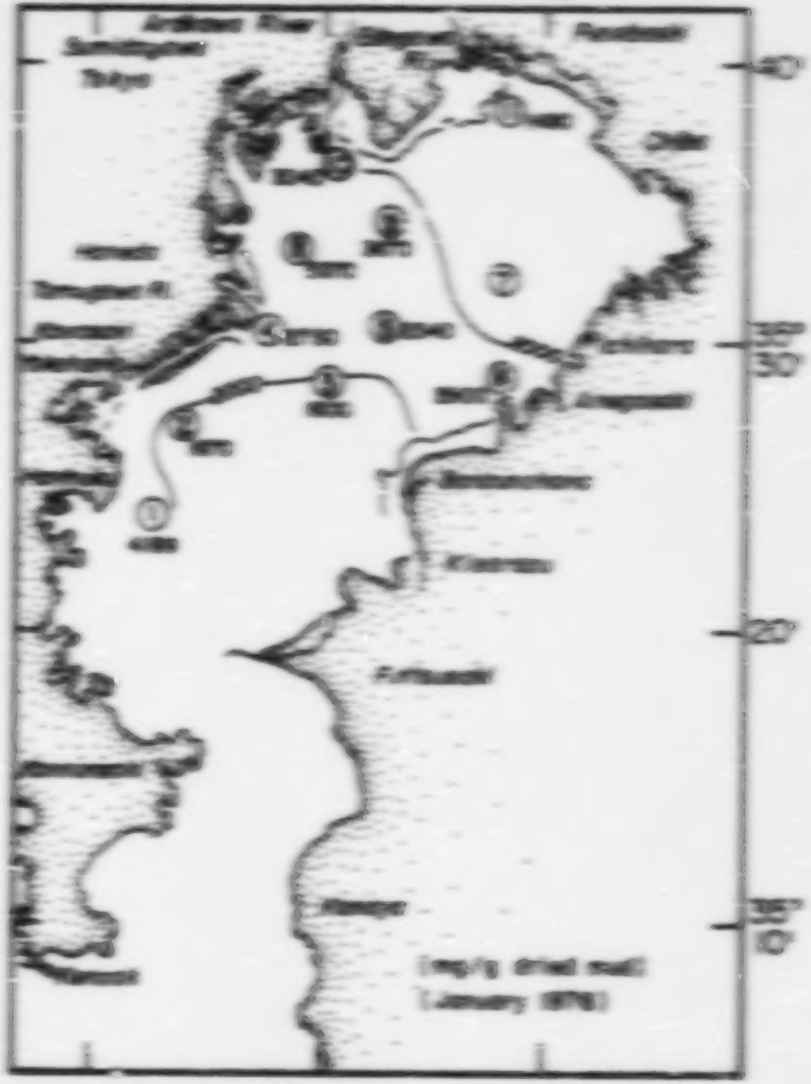
VERTICAL DISTRIBUTION OF IGNITION LOSS
Figure 16 (Silty mud layer)


40'
50'
140°
40'
35°30'
20'
35°10'
Sumidagawa R.
Tokyo
Funabashi
Chiba
Tamagawa R.
Kawasaki
Anegasaki
Kisarazu
Futtsusaki
(mg/g dried mud)
(January 1976)

HORIZONTAL DISTRIBUTION OF COD
Figure 17 (Silty mud layer)


40'
50'
140°
40'
35° 30'
20'
35° 10'
Tokyo
Chiba
(mg/g dried mud)
(January 1976)

HORIZONTAL DISTRIBUTION OF SULFIDE
Figure 18 (Silty mud layer)


40'
50'
140°
40'
35° 30'
20'
35° 10'
Tokyo
Chiba
(mg/g dried mud)
(January 1976)

HORIZONTAL DISTRIBUTION OF T-N
Figure 19 (Silty mud layer)

Tamagawa River showed concentrations of over 500 mg/kg. This area contained a large amount of nutrient salts and was considered a eutrophic area (Figure 20).

(8) BOD

High BOD densities were observed at Station No. 9 in both the silty mud layer and the 0 m layer. The values were 74.4 mg/g and 65 mg/g, respectively The gradient from No. 8 off Haneda showed progressive organic pollution of the bottom deposits. A concentration of over 30 mg/g was widely distributed in the silty mud layer, even in the middle of the Bay, except for the areas off Funabashi and Yokohama Port which showed a relatively low density of less than 20 mg/g (Figure 21). In the area on the west side of the Bay to the north of Honmokuhana, and in the middle of the Bay, the bottom deposit was polluted considerably by organic matter.

ACTUAL CONDITION OF ORGANIC POLLUTION IN TOKYO BAY AS INDICATED BY BIOLOGICAL INDEX

In the preceding section, the pollution distribution was shown in terms of actual conditions of the water quality and bottom sediments. In this section pollution is given as determined by the AGP (Algal Growth Potential) survey. This survey evaluates the potential capacity of the water in the Bay to grow algae and also used the growth of benthos as an indicator of organic pollution in bottom deposits.

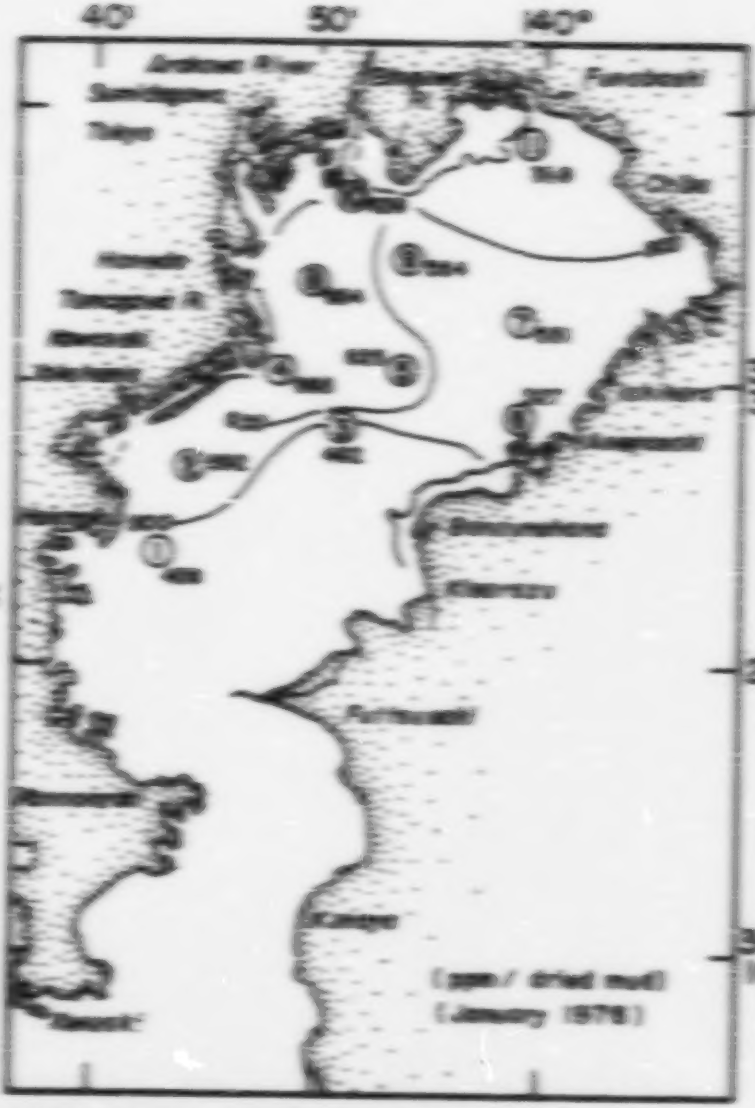
Present Condition of AGP

In December 1975, surface water was collected at 14 points in the Bay (Figure 22). Skeletonema costatum, a common Bay algae, was used in the test.

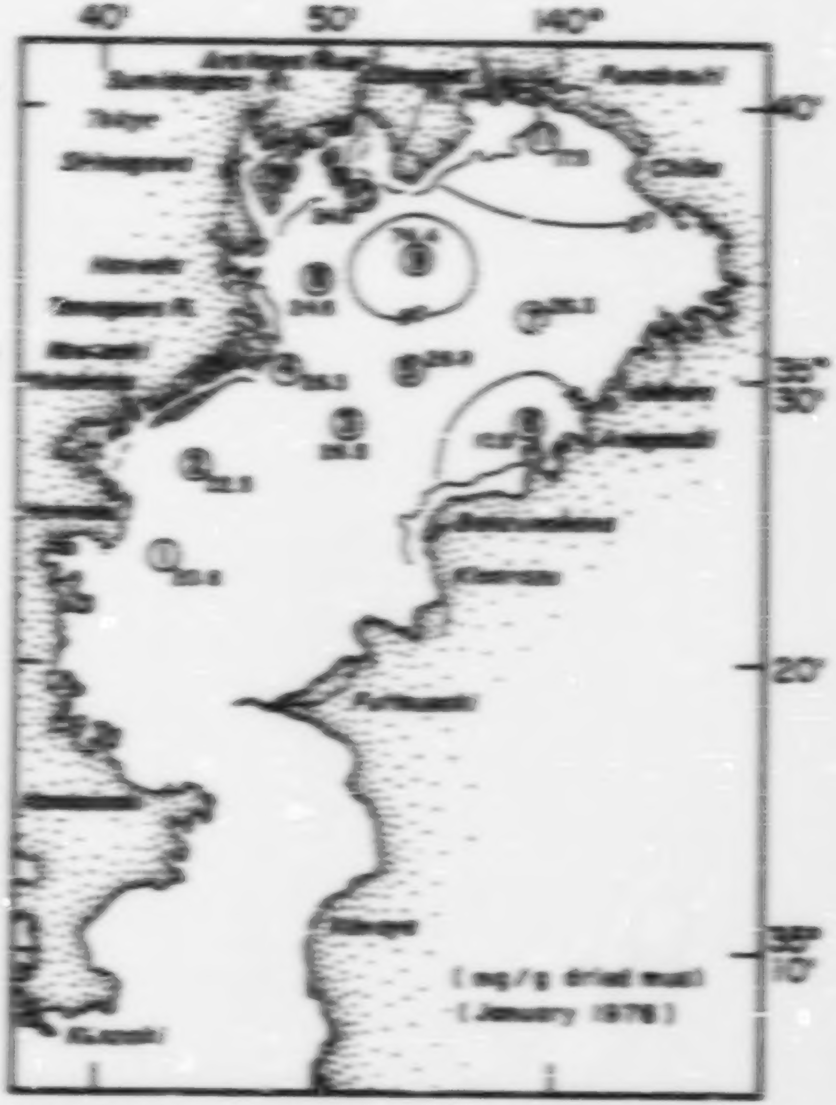
Figures 23 and 24 show that the algae tended to enter directly into the logarithmic growth period without passing through a latent period. Growth stopped after 4 to 5 days and the population began declining after 6 days in all the samples except K (Kanagawa) 4, and the control (off Arasaki Miura Peninsula). In the cases of K-4 and the control, a short latent period and less active growth were observed in the second day after the start of cultivation due to (it appears) a smaller quantity of P and N in the samples. In the cases of K-4, K-15, T (Tokyo)-8 and the control, the decline after the peak was relatively rapid.

Comparison of AGP values by sea area is shown in Table 2 and Figure 25. The Tokyo seas area showed the highest value of from 9.43×10^5 to 1.18×10^6 cells/ml, followed by 1.83×10^5 to 8.70×10^5 cells/ml in the Kanagawa sea area and 2.39×10^5 to 4.89×10^5 cells/ml in the Chiba sea area. As seen in Figure 26, the distribution of AGP resembles those of N and P.

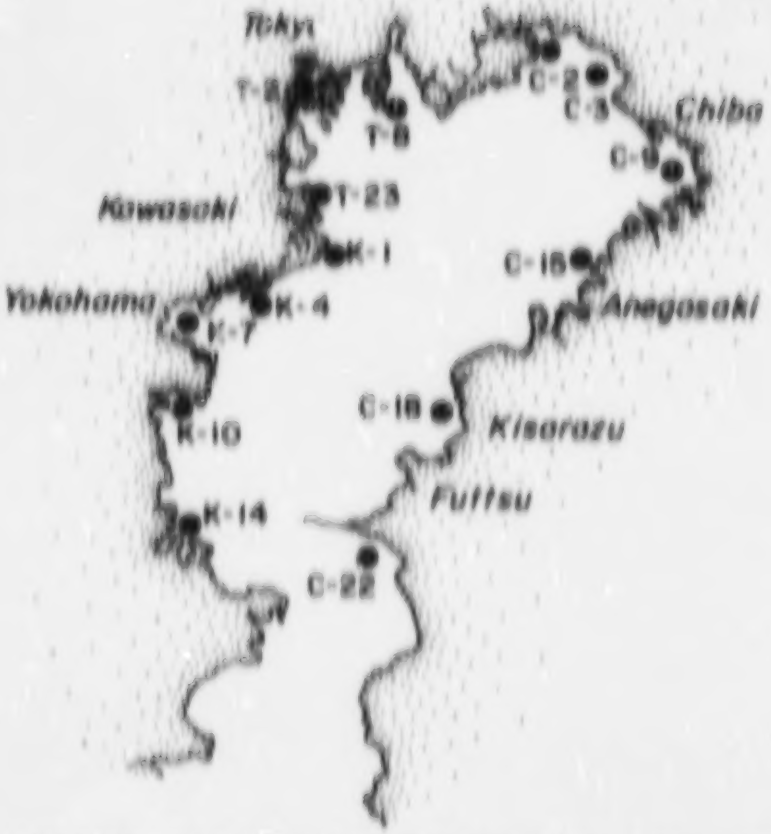
The value of T-N showed considerable changes for samples from K-7, T-23 and C-3 before and after the test and, at the remaining stations, from 50 to 60 percent of the nitrogen was consumed.


40'
50'
140°
40'
35°
30'
20'
35°
10'
(ppm / dried mud)
(January 1978)

HORIZONTAL DISTRIBUTION OF T-P
Figure 20 (Silty mud layer)


40'
50'
140°
40'
35°
30'
20'
35°
10'
(mg/g dried mud)
(January 1978)

HORIZONTAL DISTRIBUTION OF BOD
Figure 21 (Silty mud layer)


Tokyo
T-5
T-8
Kawasaki
T-23
K-1
Yokohama
K-4
K-7
K-10
K-14
C-22
C-2
C-3
Chiba
C-9
C-15
Anegasaki
C-18
Kisarazu
Futtsu

WATER SAMPLING POINTS FOR AGP TEST
Figure 22

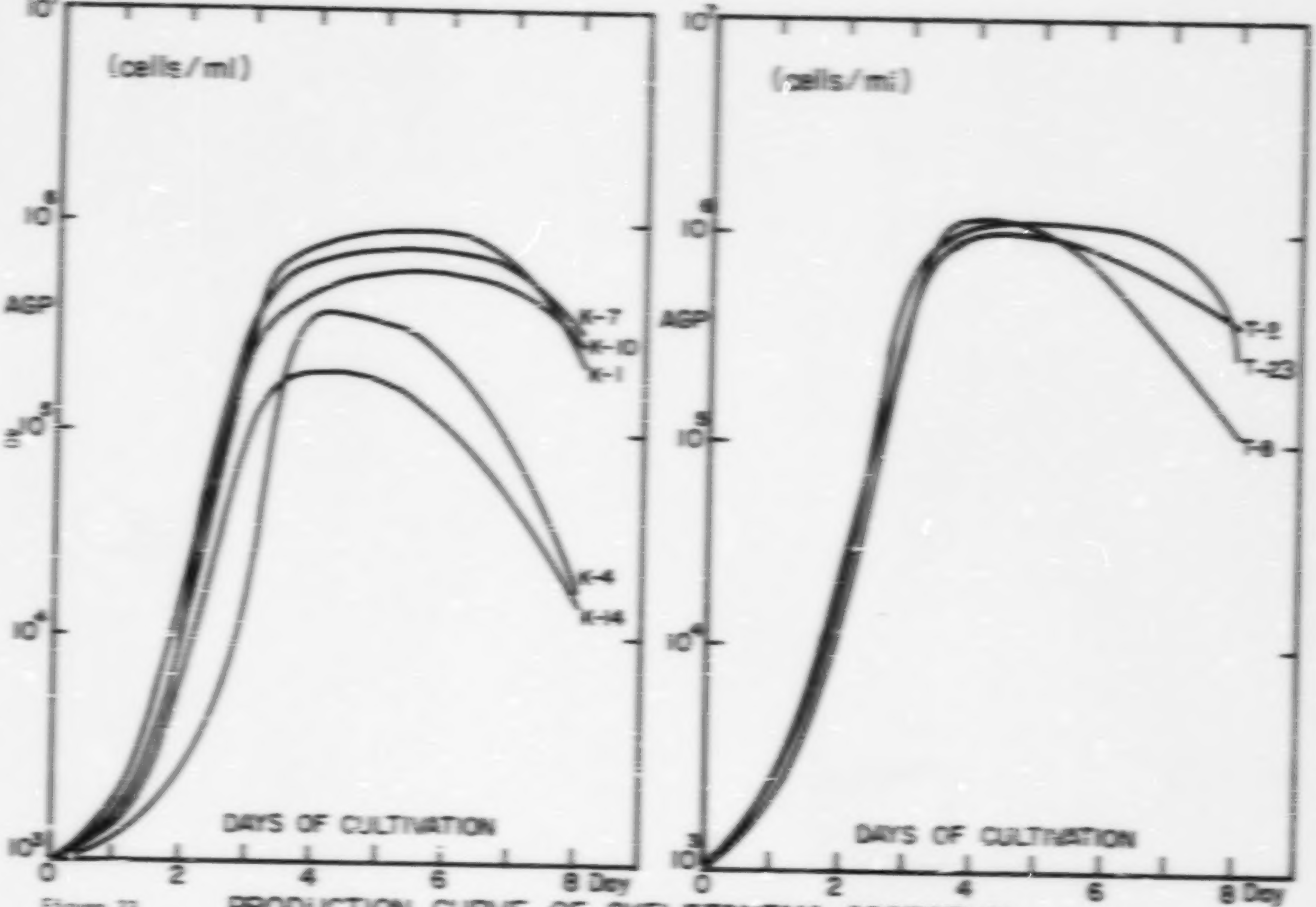


Figure 23 PRODUCTION CURVE OF SKELETONEMA COSTATUM

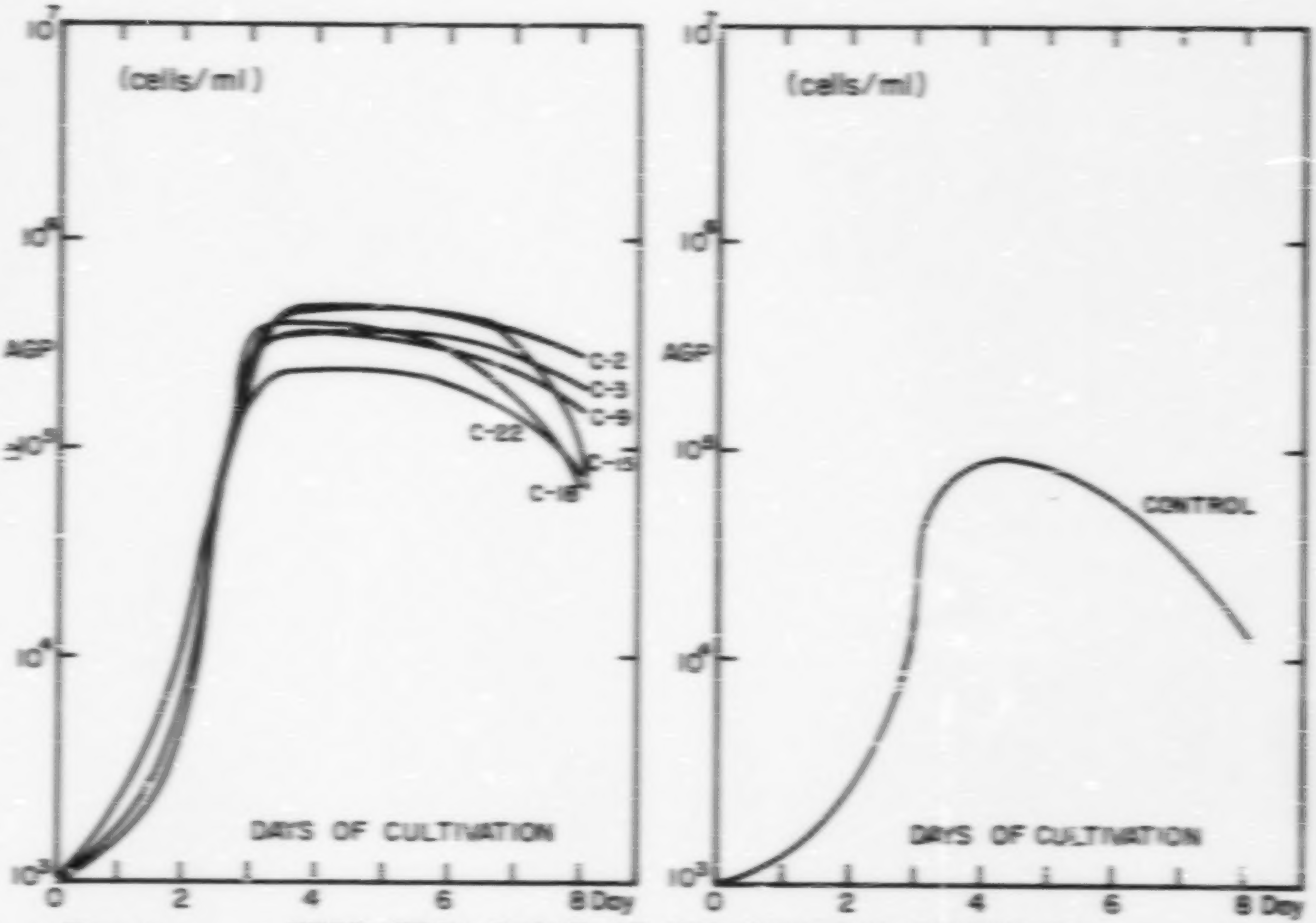


Figure 24

PRODUCTION CURVE OF SKELETONEMA COSTATUM

TABLE 2. COMPARISON OF MAXIMUM QUANTITY OF PRODUCTION BY AREA

ST.	K-14	K-10	K-7	K-4	K-1	T-23	T-2	T-8	C-2	C-3	C-9	C-15	C-18	C-22	Control
Maximum Quantity of Production	1.83 $\times 10^5$	5.60 $\times 10^5$	6.84 $\times 10^5$	4.49 $\times 10^5$	8.70 $\times 10^5$	1.06 $\times 10^6$	9.43 $\times 10^5$	1.18 $\times 10^6$	4.89 $\times 10^5$	3.59 $\times 10^5$	3.58 $\times 10^5$	4.61 $\times 10^5$	3.95 $\times 10^5$	2.39 $\times 10^5$	9.04 $\times 10^4$
1*	2.0	6.2	7.6	5.0	9.6	11.7	10.4	13.1	5.4	4.0	4.0	5.1	4.4	2.6	1.0
2*	1.0	3.1	3.7	2.5	4.8	5.8	5.1	6.4	2.7	2.0	2.0	2.5	2.2	1.3	—

*1 Shows the ratio of the maximum quantity of production at the point to the control.

*2 Shows the ratio of the maximum quantity of production at the point to K-14.

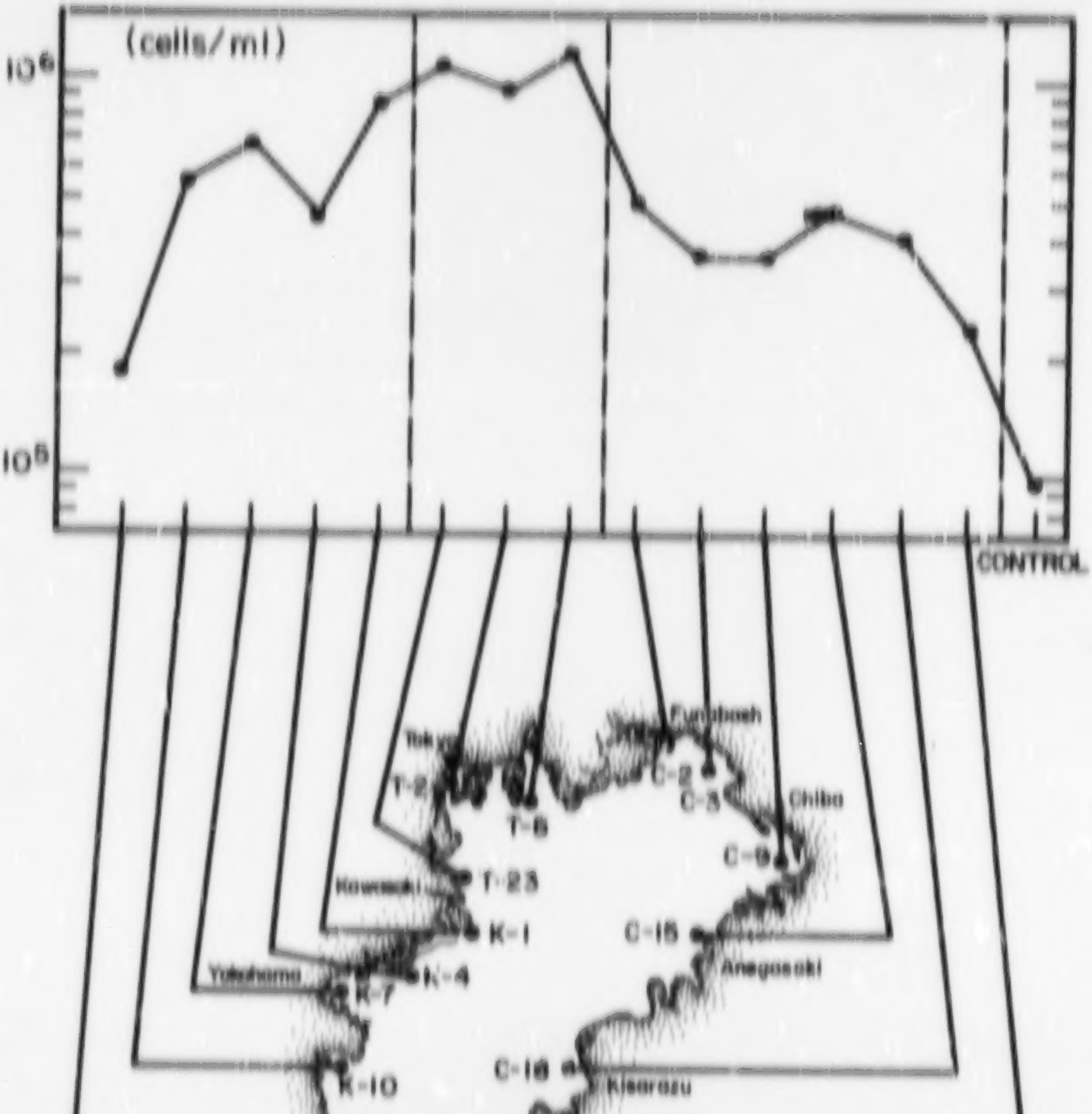
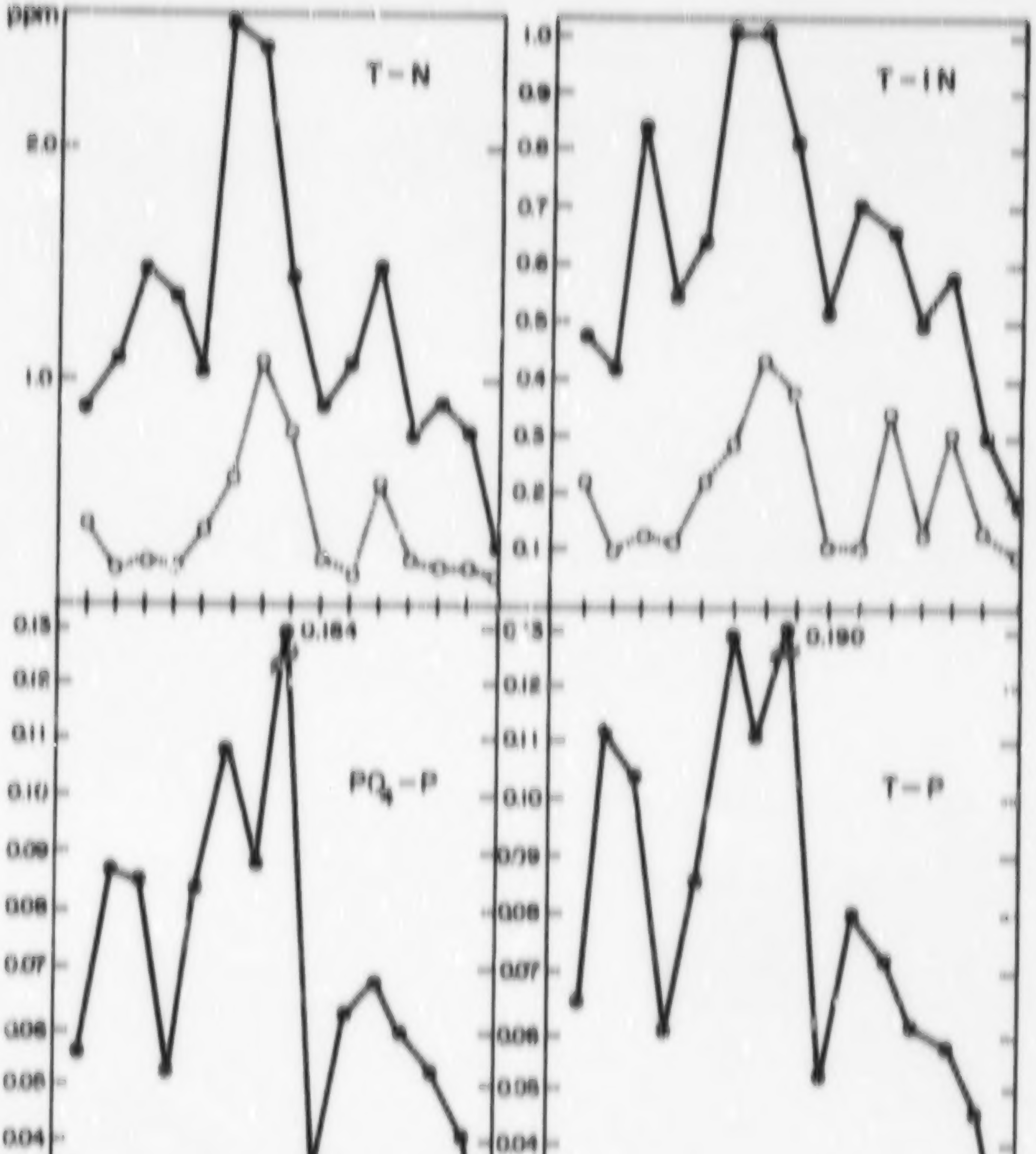


Figure 25

MAXIMUM QUANTITY OF PRODUCTION OF SKELETONEMA COSTATUM BY AREA



ANALYSIS OF WATER SAMPLE FOR AGP TEST

Figure 26

In the case of P, especially PO_4-P, the values generally decreased to 0.004 to 0.009 mg/kg after testing. This shows that *Skeletonema costatum* does not proliferate greatly once the density of PO_4-P declines to this level, in spite of the abundance of other nutrients. In other words, PO_4-P is significant as a limiting factor of AGP in Tokyo Bay.

From the above results, it was found that:

(1) AGP clearly indicates the water quality of each area, even in the Bay of Tokyo where pollution has progressed considerably, and is an effective indicator of eutrophication;

(2) Enrichment of the sea water has been progressing from Tokyo to the Kanagawa and Chiba sea areas in Tokyo Bay; and

(3) Phosphorus is a factor which limits growth of algae in Tokyo Bay.

Benthos

Because benthos has a strong tendency to live in a fixed area on the sea floor it is used as an indicator to show the general degree of pollution.

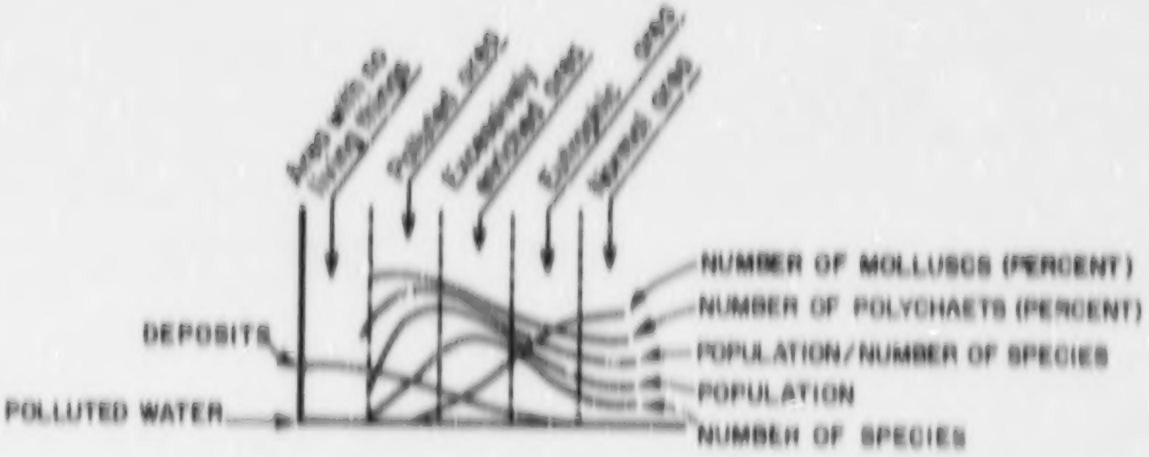
Figure 27 shows the changes in species composition of the benthos with enrichment. In non-polluted areas the benthic species composition tends to be diverse: it becomes less diverse as pollution increases. Also, as pollution increases, the ratio of the total population to the number of species (benthic index) increases because only specific kinds can survive and ultimately no benthic organisms can survive.

Therefore, the most simple standard for judging whether or not the bottom sediment is polluted is if living things are able to survive there. Figure 28 shows the abiotic areas among 11 points surveyed. The distribution is similar to that of sulfide with 1.0 mg/g (dry wt.). Judging from these two indices, it can by seen that both the coastal region and the area extending from the west side to north of the sea area in front of Anegasaki (excluding the area near to Funabashi Port) are considerably polluted.

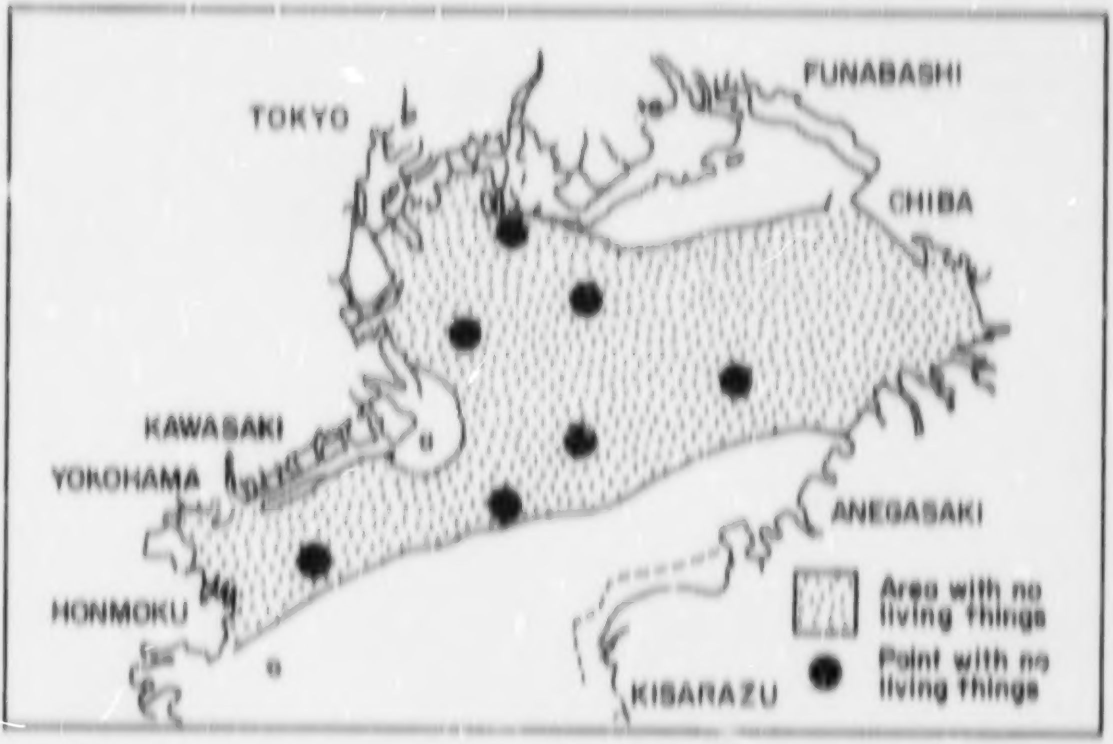
ORIGIN OF SLUDGE AND ACCUMULATION RATE OF SLUDGE IN TOKYO BAY

Origin of Sludge

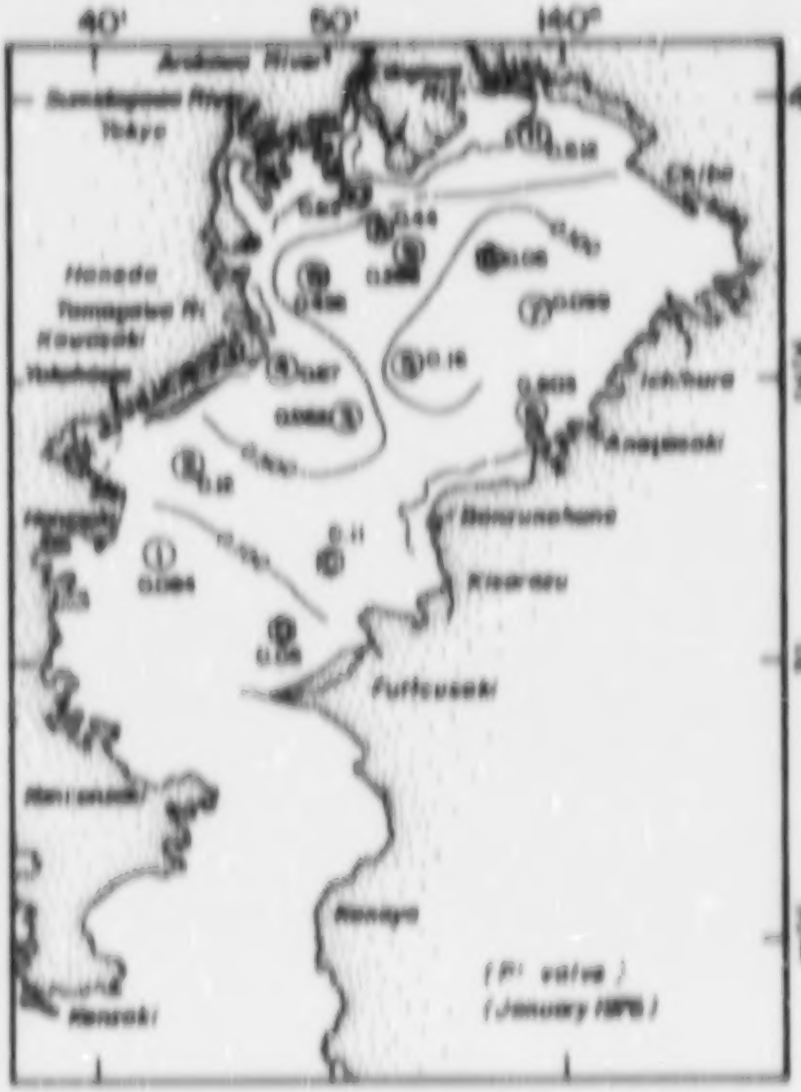
To find out the origin of organic pollution, humic acid, pheophytine and C/N ratio in the bottom were studied. Results are shown in Figures 29 through 31. Solid organic substances which settle in the bottom mud may be classified into two groups according to their origin. The first group includes marine organic substances such as dead organisms and metabolized substances from phytoplankton. The second group includes organic substances which originate on land. Generally speaking, the value of P for humic acid in the bottom deposits is higher near the mouth of the river where the inflowing river water exerts its influence and smaller at the mouth of the bay where the influence of sea water dominates and insea areas where detritus from phytoplankton accumulates.


Area with no living things
Polluted area
Excessively polluted area
Eutrophic area
Normal area
DEPOSITS
POLLUTED WATER
NUMBER OF MOLLUSCS (PERCENT)
NUMBER OF POLYCHAETS (PERCENT)
POPULATION/NUMBER OF SPECIES
POPULATION
NUMBER OF SPECIES

CHANGE IN FACIES OF BENTHOS DUE TO POLLUTION AND CLASSIFICATION OF POLLUTION (By Kitamori)
Figure 27

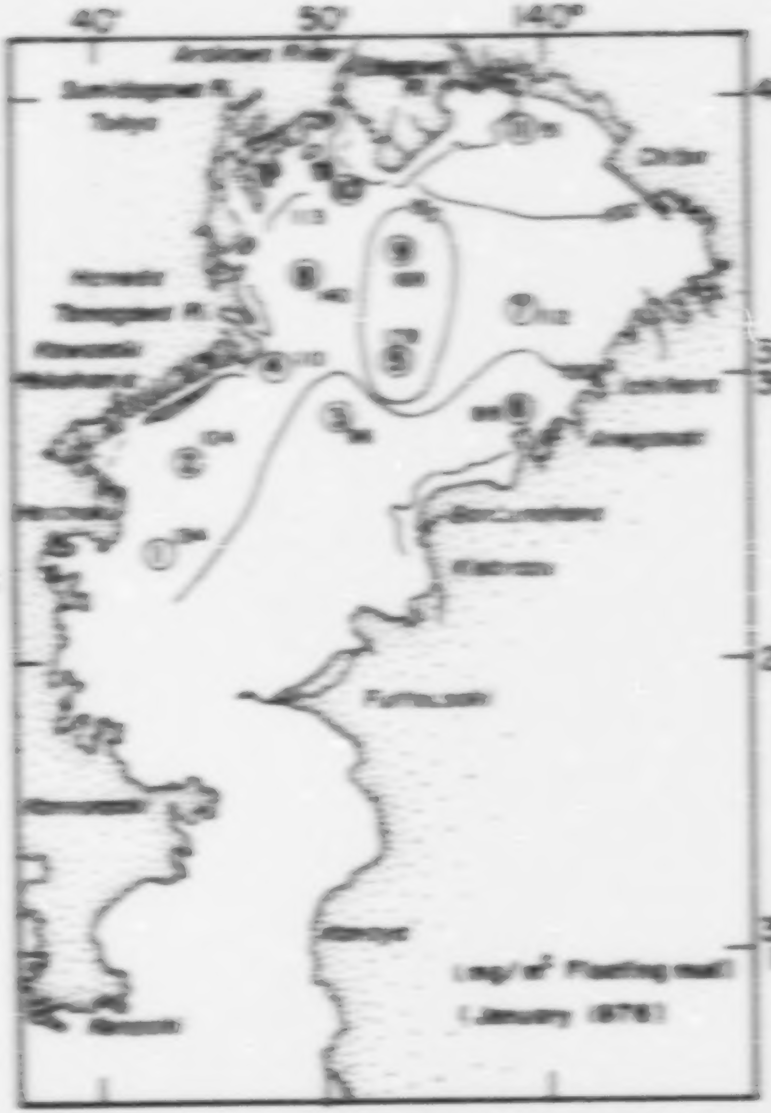

FUNABASHI
TOKYO
CHIBA
KAWASAKI
YOKOHAMA
ANEGASAKI
HONMOKU
KISARAZU
Area with no living things
Point with no living things

DISTRIBUTION OF BENTHOS (JANUARY 1976)
Figure 28

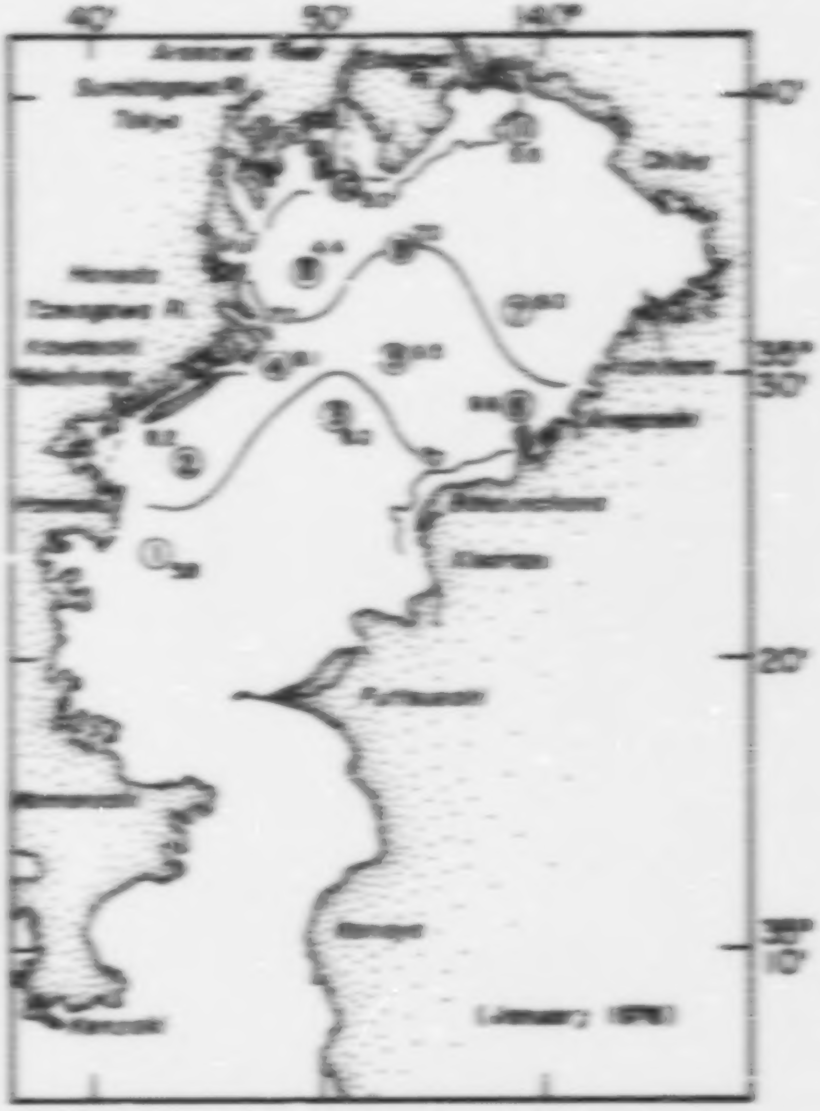

40'
50'
140°
40'
35° 30'
20'
35° 10'
Arakawa River
Sumidagawa River
Tokyo
Haneda
Tamagawa R.
Kawasaki
Yokohama
Ichihara
Anegasaki
Bannosuhana
Kisarazu
Futtsusaki
Kanaya
(P' value)

HORIZONTAL DISTRIBUTION OF HUMIC ACID
Figure 29 (0 m level)

According to Figure 29, stations 1 and D (close to the mouth of the Bay) showed small values of 0.084 and 0.050, respectively, and Stations 2 and C showed 0.120 and 0.110, respectively. This indicates that the sea water exerts a significant influence at the mouth of the Bay and a considerable part of the humic acid in the bottom deposit is sea-oriented humic acid. On the other hand, Station 4 at the mouth of Tamagawa River, Station 3 off the


40'
50'
140°
40'
35° 30'
20'
35° 10'
Tokyo
Chiba
(January 1976)

HORIZONTAL DISTRIBUTION OF PHEOPHYTINE
(0 m level)
Figure 30


40'
50'
140°
40'
35° 30'
20'
35° 10'
Tokyo
Chiba
(January 1976)

HORIZONTAL DISTRIBUTION OF C/N
(0 m level)
Figure 31

mouth, Station 10 off the mouth of Arakawa River and Station 11 off the mouth of Edogawa River showed large values from 0.512 to 0.962, indicating that the influence of the river water was very strong; a considerable part of the humic acid in the bottom deposit was land-oriented humic acid. Although small values from 0.05 to 0.160 were observed at Stations 5, 7 and 8 in the middle of the Bay, it was due, it seems, to decayed substances produced from the large amount of dead phytoplankton accumulated on the bottom.

Pheophytine is produced by decomposition of chlorophyll in phytoplankton and it has been reported that it is strongly related to the value of P for humic acid. In other words, the value of P for humic acid is small in the layer containing a large amount of pheophytine and large in the layer containing a small amount of pheophytine. This tendency was corroborated in this survey. The horizontal distribution map of Figure 30 shows values greater than 100 mg/m^2 for the area from the west side of the Bay to the deep end. The maximum value was found in the middle of the Bay, indicating a large amount of deposits mainly from phytoplankton.

There is a tendency for the ratio of organic carbon to nitrogen (C/N) to increase as accumulated organic substances decompose, and thus this ratio can be used as an indicator of the degree of decomposition of organic substances in the bottom deposits. Figure 31 shows that the degree of decomposition was small (less than 7.0) in the region north of the line connecting Haneda and Anegasaki, in the middle of the Bay (Station 3), and off the Bay off Honmoku (Station 1). The C/N ratio was large in the region extending across the Bay from Yokohama and Kawasaki to Banzunohana with values above 8.0; the maximum value of 9.5 was in the middle of the Bay, indicating that the decomposition there had progressed to a large extent.

The above findings show that: (1) The organic sludge in the area off the mouth of the Tamagawa River and the Arakawa River originated from organic substances from the land, and (2) the organic sludge in the middle of the Bay was produced from marine phytoplankton which settled on the bottom.

PCB Accumulation Rate

In this survey PCB was considered as an index and the PCB accumulation rate was measured both at Station 1 off Honmoku and Station 2 off Yokohama Port. The amount of PCB use increased every year from its beginning in 1954 to its peak in 1970. This pattern also is followed in the Keihin district. Therefore, the bottom layer with a uniform amount of PCB was considered to be a layer formed in 1954. Assuming that the sludge accumulation rate was either 1 or 2 cm/year, two different correlations were made to find the relationship between the amount of PCB used and the amount of PCB deposited in each sediment layer. Table 3 shows the results.

From the correlation coefficients shown in Table 3, it is considered that the accumulation rate was approximately 1 cm/year at Station 1 off Honmoku and approximately 2 cm/year at Station 2 off Yokohama Port. The values are close to those shown by Aizawa (1).

TABLE 3. CORRELATION BETWEEN PRODUCTION OF POS AND QUANTITY OF PCB IN THE SEDIMENT

Accumulation Speed	Measured Station Point	Regression Line	Correlation Coefficient
1 cm/year	No. 1	$y = 56.4x - 1.5$	$\gamma = 0.7$
	No. 2	$y = 6.3x + 2.9$	$\gamma = 0.1$
2 cm/year	No. 1	$y = 22.4x - 0.5$	$\gamma = 0.6$
	No. 2	$y = 6.4x + 1.2$	$\gamma = 0.7$

EFFECT OF BOTTOM DEPOSITS ON WATER QUALITY

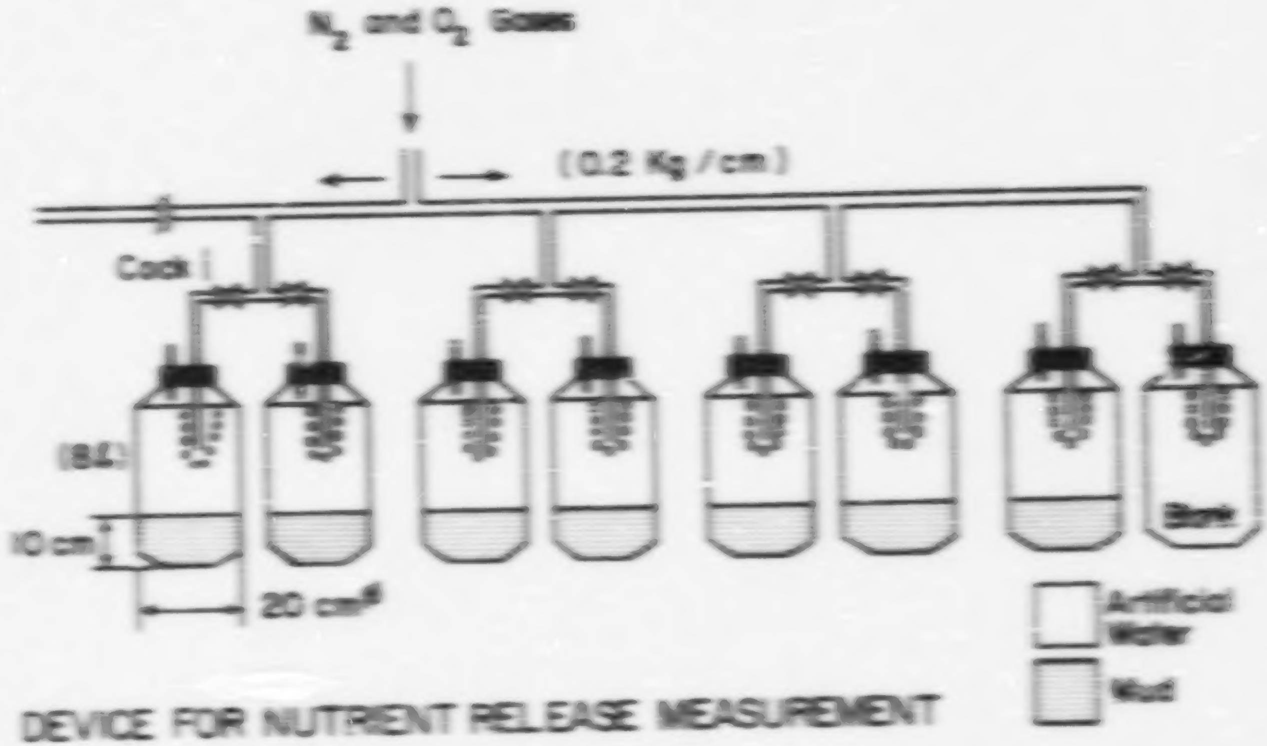
Organic matter deposited on the bottom is decomposed by bacteria. Below the surface deposits there is a limited supply of oxygen and decomposition is done by anaerobic bacteria. Anaerobic decomposition is slower in comparison to the aerobic decomposition in the water column and produces substances which impede bacterial action, further slowing down the speed of decomposition and making the bottom a storage reservoir of organic substances.

Bottom deposits are also a source of nitrogen and phosphorous. These nutrients return to the water and are used in the production of phytoplankton. This can cause a cyclical problem of increased phytoplankton production, which causes increased organic accumulation, the BOD/COD of which creates the anaerobic condition of bottom sediments, which causes release of nutrients, and so on.

This survey measured the quantity of TOC (total organic carbon), T-N (total nitrogen) and T-P (total phosphorus) released from the silty mud layer, the 0 m layer and the 2 m layer under both aerobic and anaerobic conditions. The study took place over a period of 60 days at 11 points in Tokyo Bay. Figure 32 shows the device used in the experiments. Oxygen gas (O_2) was used for the experiment under aerobic conditions and nitrogen gas (N_2) was used for the experiment under anaerobic conditions. Under aerobic conditions the concentration of T-N changed considerably at the 2 m level. T-P showed almost no change in any layer, but TOC varied considerably. Figures 33a and b show the results of the experiments for Stations 4, 5 and 6.

Under anaerobic conditions both T-N and T-P increased in concentration as time passed, and T-N increased in concentration particularly at the 2 m level. TOC did not show much variation under aerobic conditions.

The amounts of nutrients released from the bottom deposits were obtained by considering the slope of the line tangent to the release curve at the point where positive values were obtained (see Examples 1 and 2).


N_2 and O_2 Gases
(0.2 Kg/cm)
Cock
(8ℓ)
10 cm
20 cm
Blank
Artificial Water
Mud

DEVICE FOR NUTRIENT RELEASE MEASUREMENT

Figure 32

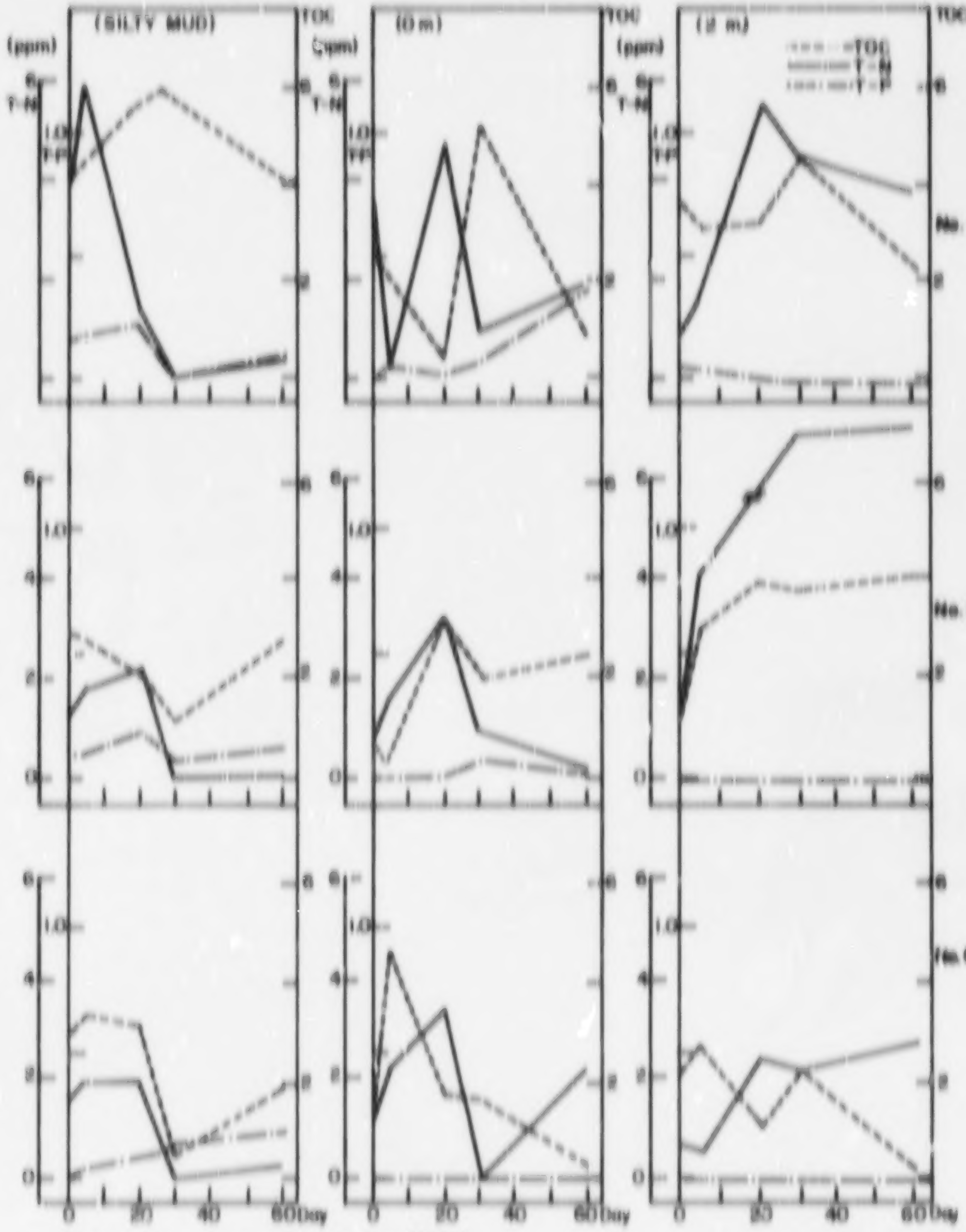


Figure 33a. Oxygen Saturation (100%)

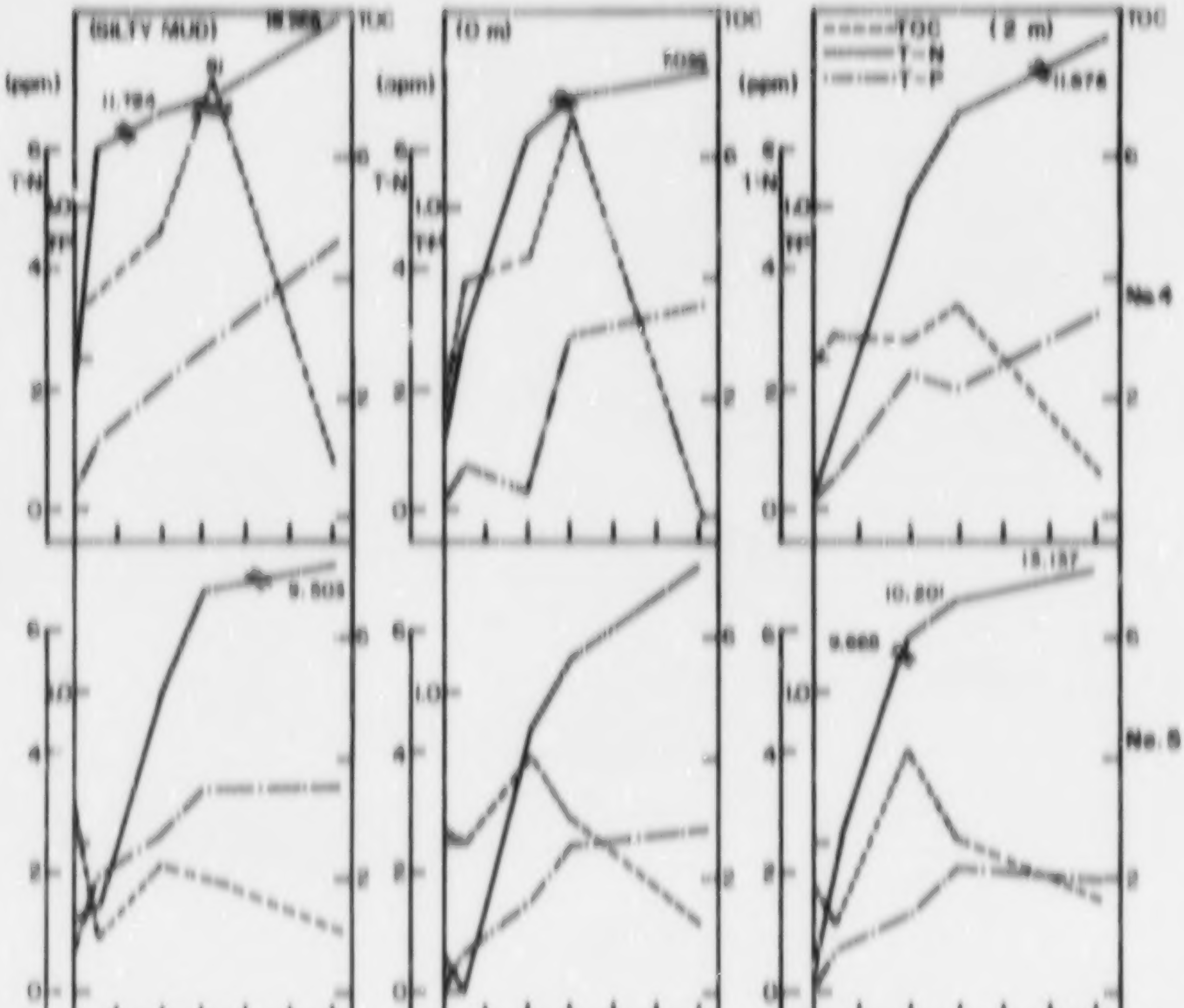
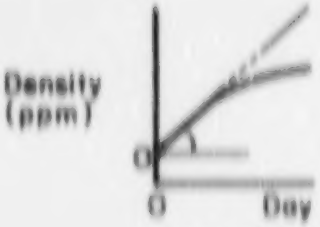
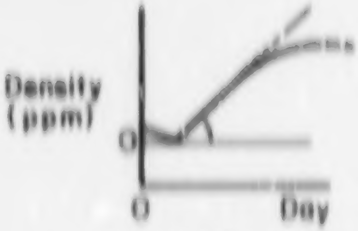


Figure 33b. Oxygen Saturation (0%)



EXAMPLE 1



EXAMPLE 2

Quantity of TOC Released

Under both anaerobic and aerobic conditions, there were large fluctuations in the release of TOC. It appears this was primarily due to the effect of biological decomposition and coagulation. The organic matter originally exists as a solid in the bottom mud. As it is decomposed by microorganisms some of it becomes soluble and is dissolved in the upper water. The organic matter released is decomposed and coagulated by microorganisms, and some of it is vaporized into the air. In order to find out the precise quantity of TOC released, it will be necessary to control decomposition, settlement and condensation by bacteria.

Quantity of Nutrient Salts (T-N, T-P) Released

Decrease in concentration of T-N was observed under aerobic conditions both in the flocculent layer and the 0 m layer. This was probably due to biological coagulation settlement and denitrification. Thus the observed decline is due to a mixture of release coagulation and denitrification. In order to find out the true quantity of T-N released further study is required. However, an estimate of the quantity of T-N released is given in Table 4. Average values from this table show aerobic conditions yielded 5.0 mg/m^2/day, 5.4 mg/m^2/day and 9.3 mg/m^2/day from the silty mud, 0 m layer and 2 m layer, respectively, and anaerobic conditions yielded 7.0 mg/m^2/day, 5.9 mg/m^2/day and 7.6 mg/m^2/day, respectively. There is not much difference in quantities released under anaerobic and aerobic conditions.

In the middle of the Bay, T-N release was over 5 mg/m^2/day and the maximum value was 23.6 mg/m^2/day at Station No. 4 off the Tamagawa River mouth. The total quantity of T-N released in the Bay was estimated to be approximately 7.0 metric tons/day, assuming that the average quantity was 7.0 mg/m^2/day and the area of the Bay was about 1000 km^2. This may be converted into COD by the equation:

$$\mathrm{COD} = \frac{19.7}{1.78} \times \alpha n \times \beta n \times N,$$

where:

αn = Conversion rate (0.230) obtained from water quality analysis of the public water area in 1974.

an = distribution ratio of plankton in the productive water area (1.0 in the sea water area).

$$COD_N = \frac{19.7}{1.78} \times 0.230 \times 1 \times 7.0 = 18 \text{ (metric tons/day)}$$

The actual value should be larger because the effect of coagulation, settlement and denitrification was included in the result and disturbance of the silty mud was not taken into consideration.

TABLE 4. QUANTITY OF T-N RELEASED

(mg/m²/day)

Layer / Condition / St. No.	Silty Mud		0 m		2 m	
	aerobic	anaerobic	aerobic	anaerobic	aerobic	anaerobic
1	0.338	3.518	2.255	4.809	17.027	6.274
2	0.860	3.737	4.755	6.338	10.810	5.988
3	1.026	2.675	0.541	2.930	7.631	8.918
4	16.390	23.599	22.690	11.274	4.771	8.281
5	3.089	1.051	5.803	9.336	18.817	13.696
6	2.733	5.701	7.147	1.210	3.928	3.376
7	5.918	6.975	5.039	8.312	11.912	6.784
8	18.225	11.019	3.007	7.484	8.504	8.886
9	4.555	4.841	5.287	5.000	12.816	12.453
10	2.096	8.503	0.508	8.408	4.090	4.245
11	0.140	5.642	2.448	0.191	2.484	5.196
average	5.034	7.024	5.407	5.936	9.345	7.645

Quantity of T-P Released

As shown in Figures 33a and b, the concentration of T-P was almost constant under aerobic conditions with very little release. About ten times as much was released under anaerobic conditions as under aerobic conditions.

Table 5 shows the quantity of T-P released. It was calculated in the same way as T-N. This table shows that under aerobic conditions the average quantity was 0.117 mg/m^2/day, 0.092 mg/m^2/day and 0.032 mg/m^2/day from the silty mud, 0 m layer and 2 m layer, respectively, and under anaerobic conditions, 0.919 mg/m^2/day, 0.502 mg/m^2/day and 0.627 mg/m^2/day were released, indicating a great variation between aerobic and anaerobic conditions.

TABLE 5. QUANTITY OF T-P RELEASED

(mg/m^2/day)

Layer / Condition / St. No.	Silty Mud		0 m		2 m	
	aerobic	anaerobic	aerobic	anaerobic	aerobic	anaerobic
1	0.061	0.153	0.042	0.191	0.064	0.612
2	0.185	1.344	0.000	0.541	0.127	0.822
3	0.185	0.287	0.081	0.605	0.032	0.548
4	0.070	1.096	0.312	0.720	0.000	0.718
5	0.025	1.675	0.006	0.790	0.000	0.867
6	0.217	1.191	0.032	0.032	0.000	0.034
7	0.306	1.108	0.248	0.287	0.000	0.331
8	0.166	0.433	0.045	0.433	0.064	1.013
9	0.142	0.701	0.000	0.389	0.032	0.656
10	0.191	1.834	0.250	1.147	0.068	1.236
11	0.047	0.287	0.000	0.382	0.008	0.057
average	0.117	0.919	0.092	0.502	0.036	0.627

The total quantity of T-P released in the Bay was estimated to be approximately 0.92 metric tons/day. This figure is based on the assumption that the average quantity was 0.92 mg/m^2/day and the area of the Bay was approximately 1000 km^2. This may be converted into COD using the equation:

$$COD_P = \frac{143}{1.78} \times \alpha p \times \beta p \times P$$

$$\alpha p = 0.585$$

$\beta p = 1$

$COD_P = \frac{143}{1.78} \times 0.585 \times 1 \times 0.92 = 43$ (metric tons/day)

The average value of both COD_N and COD_P is given as

$\frac{COD_N + COD_P}{2} = \frac{18 + 43}{2} = 31$ (metric tons/day)

This is the total quantity of the nutrient salts released.

Oxygen Consumption

Dissolved oxygen in the water is consumed by both organic matter in the water and bottom mud. Thus, the oxygen demand is great. This is particularly true in summer when the sea water is stratified and lack of oxygen becomes very serious in the bottom layer.

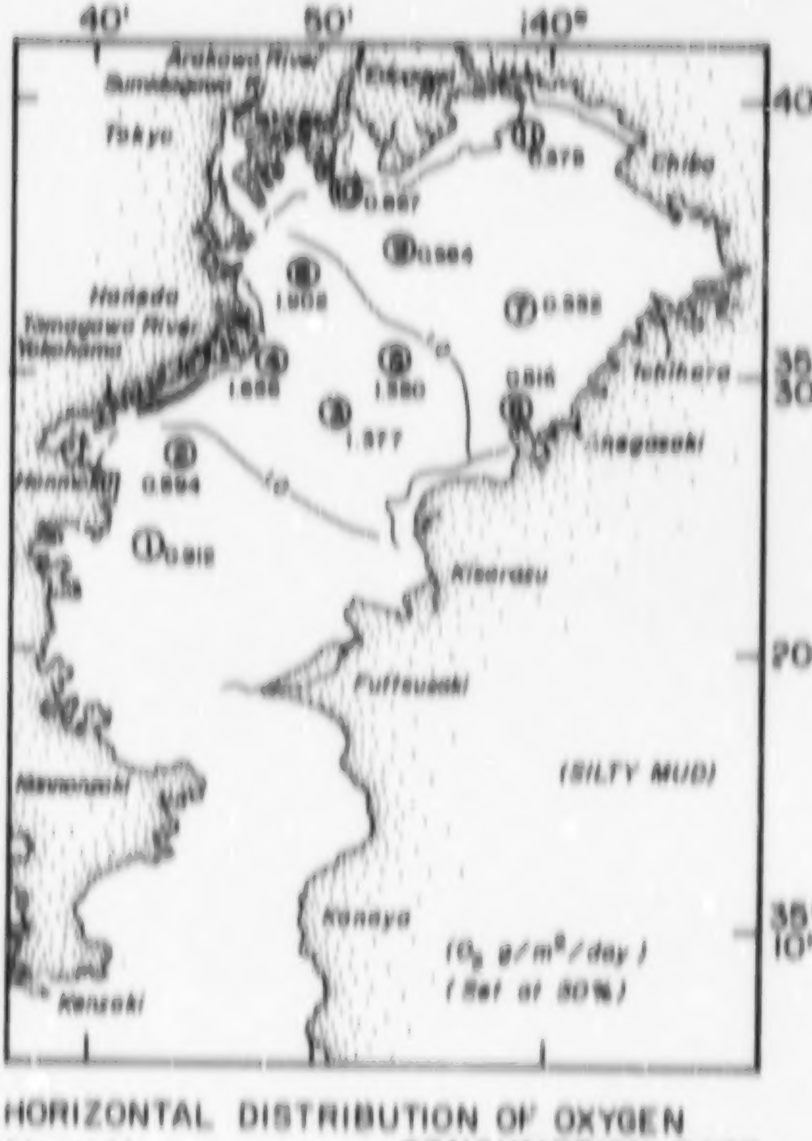
Dissolved oxygen consumption was determined in this study using a mud layer of 10 cm thickness in order to correctly represent the oxygen absorption rate in the field as shown in the report by Fair (2). Table 6 shows the results.

TABLE 6. RESULTS OF OXYGEN CONSUMPTION TEST

(O_2 g/m²/day)

Layer	Silty Mud		0 m		2 m	
Oxygen Saturation / St. No.	100%	50%	100%	50%	100%	50%
1	1.044	0.912	0.639	0.717	2.792	1.095
2	0.594	0.594	0.498	0.903	0.697	1.095
3	2.499	1.377	0.051	0.096	0.147	0.252
4	2.262	1.656	1.200	1.779	0.648	2.244
5	1.542	1.350	0.735	1.377	0.306	0.789
6	0.621	0.516	1.218	1.287	1.296	0.402
7	0.192	0.552	0.402	0.465	0.675	0.840
8	0.885	1.902	0.329	0.076	2.103	1.323
9	1.173	0.684	0.561	0.849	4.650	3.000
10	1.305	0.867	2.235	1.437	0.867	0.945
11	0.594	0.375	0.366	0.726	1.824	1.455
average	1.156	0.980	0.749	0.956	1.455	1.222

Figure 34 shows the distribution of silty mud (50%). The quantity of oxygen consumption was 1.0 g O_2/m²/day in the flocculent layer with an oxygen saturation of 50% in the area extending from the middle of the Bay to the west coast of the Bay. The maximum value was 1.9 g O_2/m²/day measured at Station No. 8 off Haneda.


40'
50'
140°
40'
35° 30'
20
35° 10'
Arakawa River
Sumidagawa R.
Edogawa
Tokyo
Chiba
Haneda
Tamagawa River
Yokohama
Ichihara
Anegasaki
Honmoku
Kisarazu
Futtsuzaki
Kannonzaki
Kanaya
Kenzaki
(SILTY MUD)
(O_2 g/m^2/day)
(Sal at 50%)
1.0
1.0
① 0.819
② 0.894
③ 1.377
④ 1.666
⑤ 1.590
⑦ 0.582
0.816
1.902
0.564
0.667
0.979

HORIZONTAL DISTRIBUTION OF OXYGEN CONSUMPTION RATE

Figure 34

The total consumption of oxygen in the Bay was estimated to be approximately 1000 metric tons/day or 600 metric tons/day in terms of COD (TOD: COD = 1.78:1) based on the assumption that the average oxygen consumption was 1.0 g O_2/m^2/day and the area of the Bay was approximately 1000 km^2.

Judging from the above result it is estimated that the silty muds consume 600 metric tons O_2/day which is equivalent to 80% of that necessary to satisfy the oxygen demand (741 metric tons/day) of materials flowing into the bay from the land on a daily basis. In spite of the fact that the silty mud contained more organic matter in comparison to other layers, it consumed a similar quantity of oxygen to that consumed by other layers. This indicates that the best method for determining oxygen consumption has not yet been established and the cause and mechanism of oxygen consumption by sludge must be determined.

SUMMARY

Eutrophication is considerably in Tokyo Bay and is caused by the large quantity of organic matter produced endogenously in the Bay as well as an equivalent amount flowing into it from the surrounding land. The sludge accumulates in the middle of the Bay. It is estimated that the quantity of accumulated sludge is approximately 400 million metric tons. This lowers the water quality by release of nutrient salts and consumption of oxygen. Moreover, the area devoid of life has been expanding.

The oxygen consumption experiment did not show a clear difference between the silty mud layer and the bottom layer (2 m layer), but it is estimated that the bottom mud consumes about 80% of the oxygen necessary to decompose all the organic substances flowing into the Bay from the land. Since the sludge consumes a large quantity of oxygen, when an overlying sludge is formed the bottom layer becomes anaerobic.

The experiment on nutrient release from the bottom muds determined that almost no phosphorus is released under aerobic conditions, but the quantity of phosphorus released increases in proportion to the quantity of phosphorus in the sediments when they are subjected to anaerobic conditions. It is therefore estimated that phosphorus equivalent to 10% of that amount flowing from the land into the Bay is released from the bottom mud at the time of formation of the sludge layer. Moreover, the AGP survey showed that the effect on the water quality of the sludge is considerable because phosphorus is the nutrient which limits phytoplankton productivity in Tokyo day.

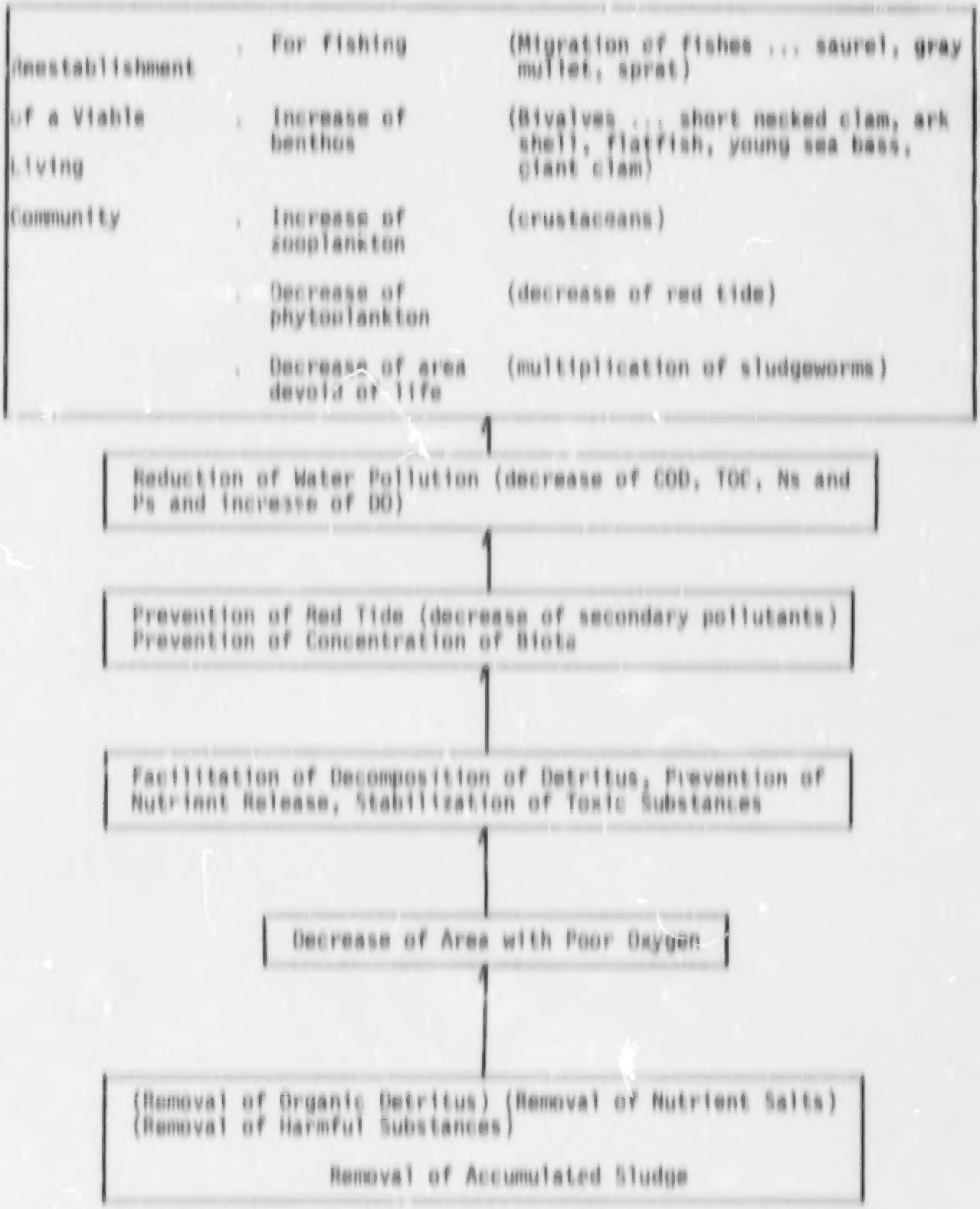
It is expected that by removing the accumulated sludge the environment can be improved through the cycle as shown in Figure 35. If the silty mud layer is removed COD will be less than 30 mg/g and sulfide less than 0.3 mg/g (up to the 0.5 m layer at Station 4 and up to 1 m layer at Station 10) to make the bottom mud either clay or silt. Here the water quality should be taken into consideration. According to Yoshida's table (3) which classifies sea area by nutrient level and bottom characteristic, a polluted water area can become a more desirable enriched water area where benthos and crustaceans may increase in kind and number.

Finally, problems which need work are listed below.

(1) Mechanisms of bottom pollution

More work is required on dispersal of organic sludge and on oxygen consumption rates. It will be necessary to research suspended solids and the dispersion of bottom mud.

Figure 35. Flow Chart for Purification of Bottom Deposits in Tokyo Bay


Reestablishment of a Viable Living Community
. For fishing (Migration of fishes ... saurel, gray mullet, sprat)
. Increase of benthos (Bivalves ... short necked clam, ark shell, flatfish, young sea bass, giant clam)
. Increase of zooplankton (crustaceans)
. Decrease of phytoplankton (decrease of red tide)
. Decrease of area devoid of life (multiplication of sludgeworms)
Reduction of Water Pollution (decrease of COD, TOC, Ns and Ps and increase of DO)
Prevention of Red Tide (decrease of secondary pollutants)
Prevention of Concentration of Biota
Facilitation of Decomposition of Detritus, Prevention of Nutrient Release, Stabilization of Toxic Substances
Decrease of Area with Poor Oxygen
(Removal of Organic Detritus) (Removal of Nutrient Salts)
(Removal of Harmful Substances)
Removal of Accumulated Sludge

(2) Effect of sludge

The general effect of bottom mud and water quality on the ecosystem was determined. To define this in more detail it is necessary to find out 1) the effect on eutrophication (in the form of AGP) of nutrients contained in the mud, 2) the effect of bottom sediments on living organisms through bioassay and 3) oxygen consumption of mud and sludge to be removed.

(3) Technology of pollution abatement

Dredge reclamation, treatment at the original place and ocean dumping of dredged materials should be comprehensively evaluated from the standpoints of environment, technology and economics. It is also necessary to develop a technology to facilitate settling of dredged materials at the dredge and dump sites.

(4) Effect of bottom purification

It is necessary to quantitatively determine the effect of improving water quality via bottom purification by using a model in which the cycling of substances and the exchange of sea water are taken into consideration. At the same time the model should account for the effect of bottom purification on the recovery of the ecosystem.

REFERENCES

1) Takako Aizawa, "Vertical Distribution of Cadmium, Total Mercury and PCB in the Bottom of Tokyo Bay," Bottom Pollution Improvement Countermeasure Survey Research Paper, 1974.
2) G. M. Fair, "Water Supply and Waste Water Disposal."
3) Yohichi Yoshida, et al., "Change in Production of Living Things in the Lower Production Stage," Eutrophication and Cultivation of Marine Products in the Water Area, April, 1973, Japanese Fishery Society, Koseisha-Koseikaku.

AN EXPERIMENT IN REMOVAL OF ORGANICALLY POLLUTED BOTTOM MUD FROM THE SETO INLAND SEA

Akio Murakami*
Nansei Regional Fisheries
Research Laboratory
Fisheries Agency

ABSTRACT

Since the 1950's organic pollution of bottom mud in the Seto Inland Sea has been observed near the pulp mill wastewater drain. By the 1970's the polluted area had expanded (Figure 3). The Fisheries Agency examined this problem from August to October in 1974 (Figures 1, 2, Tables 1, 2). According to the Agency, concentrated areas of polluted mud were observed in Osaka Bay, the eastern part of Hiuchi Nada, Hiroshima Bay, Beppu Bay and some other areas (Figures 4-9, Tables 3, 4). The upper 20 to 30 cm of the bottom mud was heavily polluted. The volume of organically polluted bottom mud (COD >40 mg/g) is 10^6 m^3 in the Seto Inland Sea, 85% of which exists in Hiroshima Bay (Figures 10, 11, Table 5).

The red tide in the Seto Inland Sea became harmful in the 1960's and this is related to bottom pollution. Eutrophication in inshore waters contributes to the outbreak of the red tide, and the organically polluted bottom mud contributes to the eutrophication. Since 1973, the Fisheries Agency has been removing the polluted mud to reduce the eutrophication.

In this project the organically polluted bottom mud is dredged using a variable volume volute pump to prevent the diffusion of mud. The dredged mud is transferred to the treatment pontoon by a suction pipe. Then the mud is treated through the processes of coagulation, sedimentation and dehydration. Activated charcoal is used to filter the decant water which is then discharged to the sea. The settled sludge is hardened with cement and then dumped in a reserved land area (Figure 13).

---

*7782-9 Maruishi, Ohno-cho, Saeki-gun, Hiroshima-ken 739-04, Japan

There are still some problems to be solved in our experiment. Work needs to be done on the method of removing newly deposited materials from the surface of polluted bottom muds.

INTRODUCTION

The increase of organic pollution of bottom mud in the Seto Inland Sea has spread from stagnant regions, where it has been exacerbated by the lack of dissolved oxygen at the bottom layer and eutrophication due to nutrients released from the sediment. The marine ecosystem has been severely damaged. This paper discusses the mechanism and the influence of organic pollution of bottom mud, reports on the organic pollution of the bottom mud in the Seto Inland Sea in 1974, and describes the Fisheries Agency project for abating this pollution by removal of the bottom mud.

MECHANISM AND INFLUENCE OF ORGANIC POLLUTION OF BOTTOM MUD

Solid organic materials which end up in bottom sediments are produced both in the sea and discharged from the land. Once discharged pollutants have grown beyond the capability of the sea to process them, the sea-water becomes polluted. This occurs first in the neighborhood of the drainage outfalls when solid organic materials in the waste waters (or detritus produced in the sea) settle to the bottom near the outfall. Bottom mud pollution lags behind water pollution, and is restricted to a more narrow area. Water pollution is ephemeral, while a history of sediment pollution is recorded continually by layers of deposited material. The area polluted is dependent on the quantity of pollutant and the prevailing currents. In the case of bottom mud pollution, particulate materials sink mostly at the center of eddy currents or in stagnant regions. The particle size of sediment in such regions is very small, and the surface layer is covered with fine silt commonly called "Hedoro" in Japan.

In summer a thermocline is developed because the current is generally slow in inshore waters except at the mouth of the bay and in the strait. At that time the surface water is not mixed with bottom water and occasionally the temperature difference between the two is 10°C or more in July and August. In autumn, the thermocline disappears due to the cooling of the surface water and mixing of the two layers occurs. Moreover, they may suddenly be mixed in shallow waters by strong winds such as a typhoon. In such cases, silt at the sediment-water interface is mixed into the overlying water and may transfer to another stagnant region where it settles again. Therefore, polluted bottom mud often is found far from the drainage outfalls where the contamination originates. In the Seto Inland Sea the polluted sediments in the central part of Harima Nada, the eastern part of Hiuchi Nada, and the southern part of Suou Nada have resulted from such processes.

Organic pollution in the bottom mud acts as a secondary source of water pollution. Although nitrogen and phosphorus do not affect eutrophication as long as they are combined with the mud, when released by bacterial or chemical action they can be transferred into bottom water and from there move through mixing into the upper waters where they contribute to eutrophic conditions.

Particulate organic nitrogen (PON) and particulate organic phosphorus (POP) are mineralized both in suspension and in sediments. The amount of particulate organic materials existing in the bottom and the surface sediments is related to the degree of eutrophication of the marine environment. The bottom mud polluted by the waste water from pulp mills generally does not contain many nutrients, but it consumes dissolved oxygen in sea water as it is oxidized. This results in a lack of dissolved oxygen in bottom waters (especially in the summer thermocline stage), and many benthic organisms are killed. Since the organic compounds (vitamins, etc.) and heavy metals (iron and manganese, etc.) stimulate the growth of flagellates, an important countermeasure against the red tide is to prevent the release of such substances from the bottom mud.

STATUS OF SEDIMENT ORGANIC POLLUTION IN THE SETO INLAND SEA

The Seto Inland Sea is the largest inshore body of water in Japan. It is about 400 km in length and 21,827 km^2 in area and its average depth is 37.3 m. Depth in the straits is more than 100 m. There are about 800 islands and many bays, Nadas and straits. Two branches of the Kuroshio current enter this area via Bingo and Kii channel, and the Tsushima current also enters via the Shimonoseki strait. Many rivers flow into this area. Consequently, the water in the Seto Inland Sea is slowly transferred from west to east and there are many stagnant regions in it. Thus the exchange of water with the ocean is very poor. Marine pollution has been observed since the 1950's, and became serious around 1973 or 1974. Due to the regulation of pollution loads by the "Seto Inland Sea Conservation Law" in 1973 the situation is improving.

The Fisheries Agency currently is studying sediment pollution in the inshore waters of Japan. They carried out research in the Seto Inland Sea from September to October, 1974 and in Ise and Mikawa Bay, Hibiki Nada and Hakata Bay from August to September, 1975 and also in Omura Bay and Ofunato Bay in July, 1976 (1). The volume of polluted bottom mud has been estimated from the results. The investigations were carried out by the Fuyo Ocean Development, Co. at 160 stations in the Seto Inland Sea and another 924 stations in 26 heavily populated areas. The sampling and analyzing methods are shown in Tables 1 and 2. Mud samples were collected with the core sampler shown in Figure 1 and frozen until the analytical work was performed. The echo sounder investigation was carried out at 160 stations with a Sonicator Model RS-72. Two ultrasonic frequencies (400 kHz and 30 kHz) were used and the thickness of the mud-water layer was determined from the difference of the return time between the 400 kHz and 30 kHz waves. The sea regions in the Seto Inland Sea and the stations are shown in Figure 2.

Many local sediment studies relating to fisheries impacts have been carried out in the Seto Inland Sea. But the investigations carried out by our laboratory (2) in December 1970, August 1971, October 1972 and May 1973 were the first time the inland sea had been investigated in entirety.

Sediment from the upper 3 cm layer was collected with an Ekman-Birge dredge at 100 stations and analyzed for COD, Ignition Loss (IL)* and total-S.

---

* ash-free dry weight

TABLE 1. STANDARDIZED SAMPLING PROCEDURE FOR BOTTOM MUD

Sampling Method	Sampling Length of Mud (cm)	Layer of Mud for Analysis (cm)				
		0 - 15	15 - 20	30 - 35	5 Between Bottom and 35 cm Layer	5 From Bottom
Ekman-Birge	<10	X	—	—	—	—
Smith-McIntyre	10-20	X	—	—	—	X
Core Sampler	<20	X	—	—	—	X
	20-35	X	X	—	—	X
	35-50	X	X	X	—	X
	>50	X	X	X	X	X
Item of Analysis	COD Water Cont.	X	X	X	X	X
	II. Total-S. P. N. Silt Comp.	X	—	—	—	—

TABLE 2. METHOD OF ANALYSIS OF BOTTOM MUD

Items	Methods	Items	Methods
Size Composition	Screen ca. 50 g mud with water by 32 and 150 mesh sieves, dry the residue at 110°C, weigh after cooling.	COD	alkaline potassium permanganate, iodometric titration
		IL	700°C 2 hrs.
	(division of bottom mud)	Total-S	detection tube
	Coarse Sand Gravel: not pass through 32 mesh (dia. -0.5 mm)	P	Strickland and Parson's "A Practical Handbook of Seawater Analysis" (1968)
	Sand: not pass through 150 mesh (dia. 0.125-0.5 mm)	N	CN - Corder
	Mud: pass through 150 mesh (dia. -0.125 mm)		

IL was determined by burning the mud at 800°C until constant weight was achieved. Total-S content was determined using the following method: The mud was steam distilled and hydrogen sulfide from it was absorbed by a solution of zinc-acetate titrated with 1/100 N iodine. Results showed that the degree of bottom mud pollution changes seasonally--it increases during summer and autumn, and decreases during winter and spring. The maximum values of COD, IL and total-S are often found in autumn. The distribution of COD, IL and total-S for October, 1972 is shown in Figure 3. It is obvious that the mud is seriously polluted in Osaka Bay, the northern coast of Harima Nada, Hiuchi Nada, Hiroshima Bay, the southern area of Suho Nada and in Beppu Bay. The higher values (e.g., COD 30 mg/g, IL 14%, total-S 0.5 mg/g) are also observed at the central part of Harima Nada, Bingo Nada, Hiroshima Bay and the mouth of Beppu Bay during summer and autumn. COD and IL values do not exhibit the seasonal variation similar to the total-S values. Although the regions where total-S content exceeds 0.5 mg/g are observed everywhere during summer and autumn, the values decrease below 0.2 mg/g in spring at almost all stations. The nearest station to the coast is 2 or 3 km offshore, but there are regions of about 35 mg/g COD and 1.0 mg/g total-S offshore of Kure Bay and the central part of Osaka Bay. The area of heavily polluted sediment offshore had already increased by the time of the survey.

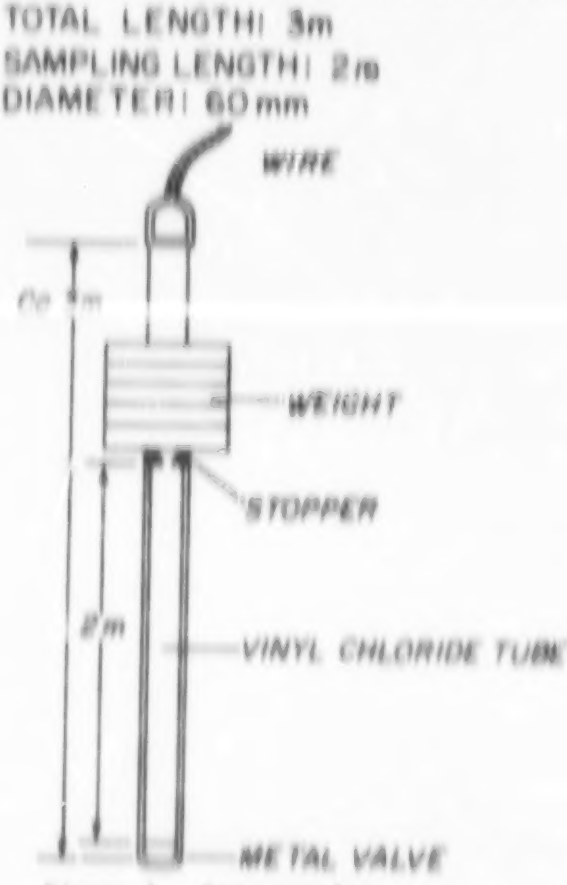


Figure 1. Core sampler.

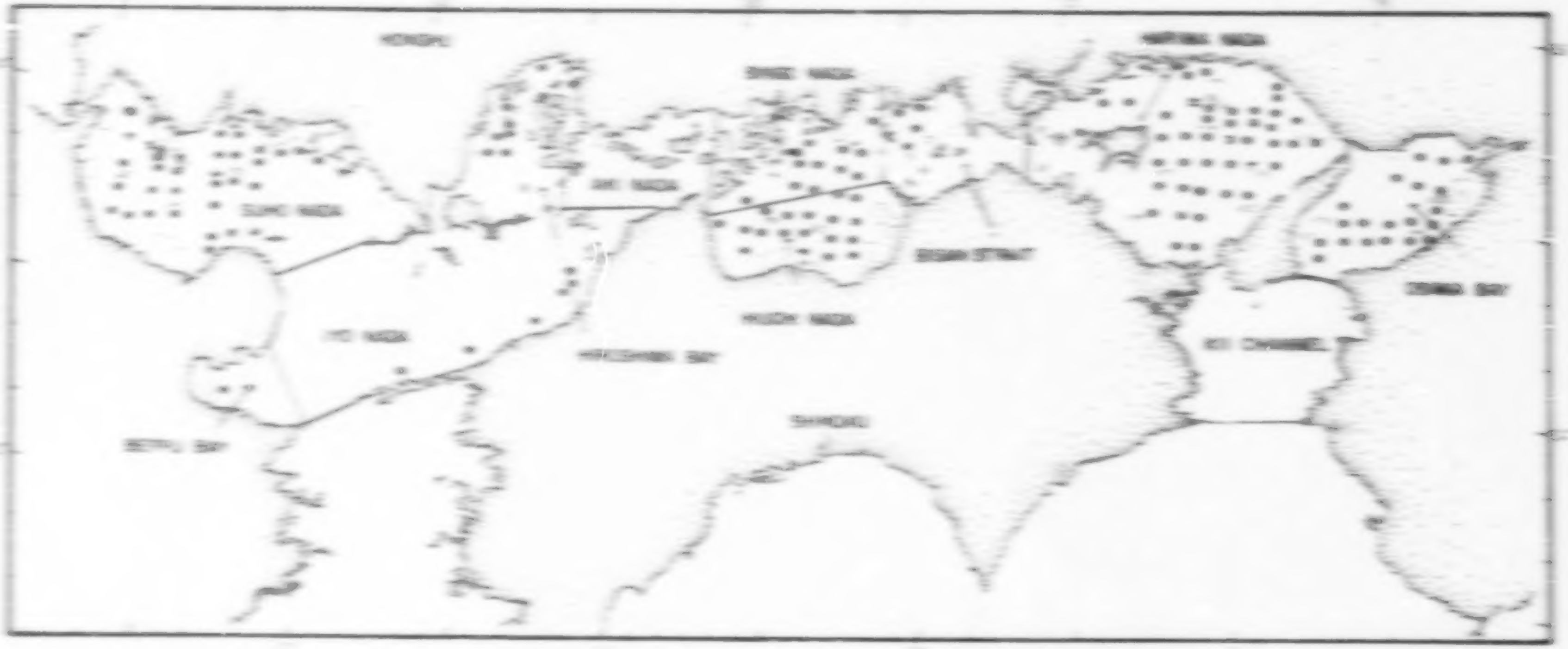

HONSHU
SUWO NADA
AKI NADA
BINGO NADA
HARIMA NADA
IYO NADA
HIROSHIMA BAY
HIUCHI NADA
BISAN STRAIT
OSAKA BAY
KII CHANNEL
BEPPU BAY
SHIKOKU

Figure 1. Sampling stations of bottom mud in the Seto Inland Sea. Aug–Oct 1974.

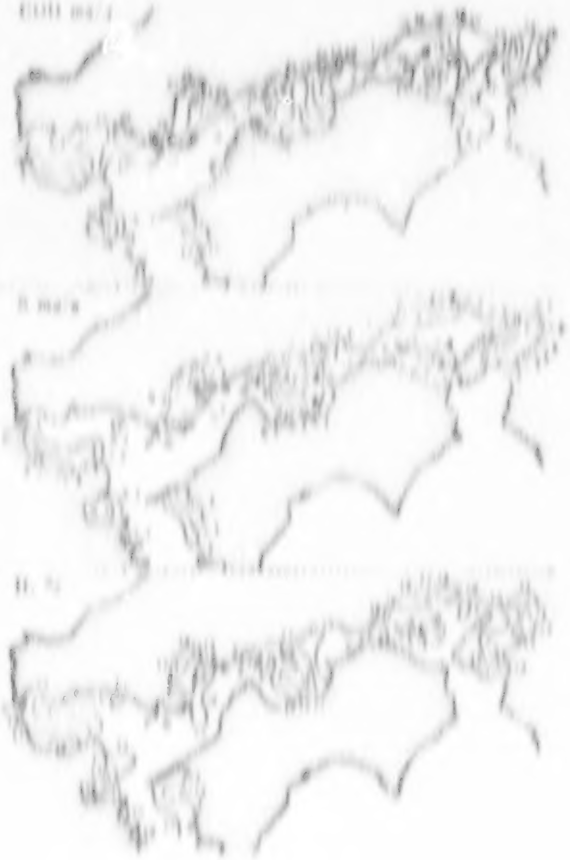


Figure 3. Bottom mud qualities (COD, Total-S, Ignition Loss) in the Seto Inland Sea, October 1972.

The 1974 investigation was carried out from September to October during the season when pollution is most severe. More than 1,000 stations from 2-300 m off the coast were examined. Nutrient values as well as the usual measurements were determined. The results are shown in Figures 4-9. The maximum, minimum and average values of measurements in several regions are shown in Table 3. As shown in these figures and the table there are many heavily polluted regions. Although these are the same regions as in former research projects, the maximum values of COD, IL and total-S have increased. High nitrogen and phosphorus contents (3.0 mg/g and 1.5 mg/g, respectively) are presumed to be due to interstitial water containing dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphorus (DIP). According to Ukita et al., (3), the mud columns collected off Iwakuni, Mitajiri, Tokuyama and Ube, are black at the surface, but gradually fade with depth. Generally, nitrogen and phosphorus in the upper 5 cm layer are respectively 2 to 3 times and 1.5 to 2 times greater than those below the 20 cm layer. Nitrogen and phosphorus contents are especially high in Mitaziri Bay which is polluted by

TABLE 3. QUALITY OF BOTTOM MATERIALS FROM SEVERAL SEA REGIONS IN THE SETO INLAND SEA, AUG.-OCT., 1974

Item \ Sea Region		Suwo Nada	Hiroshima Bay	Iyo Nada	Bingo Nada	Hiuchi Nada	Bisan Strait	Harima Nada	Osaka Bay	Kii Suido	Inner Beppu Bay
COD (mg/g)	Max.	26.2	40.7	7.8	23.2	20.3	12.9	23.1	21.8	12.9	36.6
	Av.	14.3	23.5	4.2	10.3	8.3	7.8	12.1	6.6	10.0	14.1
	Min.	4.3	12.8	1.3	2.5	2.0	2.4	1.2	1.3	6.4	2.3
IL (%)	Max.	16.5	17.8	12.8	14.8	13.8	12.0	13.8	14.0	8.4	18.8
	Av.	11.0	12.8	8.5	11.0	10.7	8.6	9.6	10.0	6.6	14.2
	Min.	3.1	9.2	4.7	8.4	6.8	2.7	2.6	3.5	3.6	1.8
Total-S (mg/g)	Max.	1.0	0.6	0.1	0.9	0.4	0.8	1.2	1.3	0.3	1.4
	Av.	0.3	0.2	0.2	0.4	0.2	0.3	0.3	0.5	0.1	0.3
	Min.	0.01	0.04	0.01	0.1	0.04	0.01	0.01	0.01	0.02	0.02
P (mg/g)	Max.	1.1	0.9	1.2	0.9	0.9	0.9	0.9	0.9	0.8	1.5
	Av.	0.6	0.7	0.6	0.7	0.6	0.5	0.6	0.5	0.7	0.9
	Min.	0.2	0.6	0.4	0.5	0.4	0.2	0.2	0.2	0.7	0.5
N (mg/g)	Max.	2.5	2.8	1.2	2.5	2.3	1.9	2.5	2.0	1.3	3.0
	Av.	1.6	2.3	0.7	1.9	1.7	1.3	1.7	1.2	1.1	2.2
	Min.	0.4	2.0	0.3	1.4	0.9	0.3	0.4	0.4	0.6	0.4
Composition of Mud (%)	Max.	100.0	99.4	77.5	98.7	98.9	95.7	98.5	99.0	98.2	99.9
	Av.	75.9	97.6	38.3	97.2	94.6	67.3	87.6	53.1	88.3	91.8
	Min.	10.3	94.2	1.1	86.2	63.2	12.0	2.7	9.8	77.1	14.5

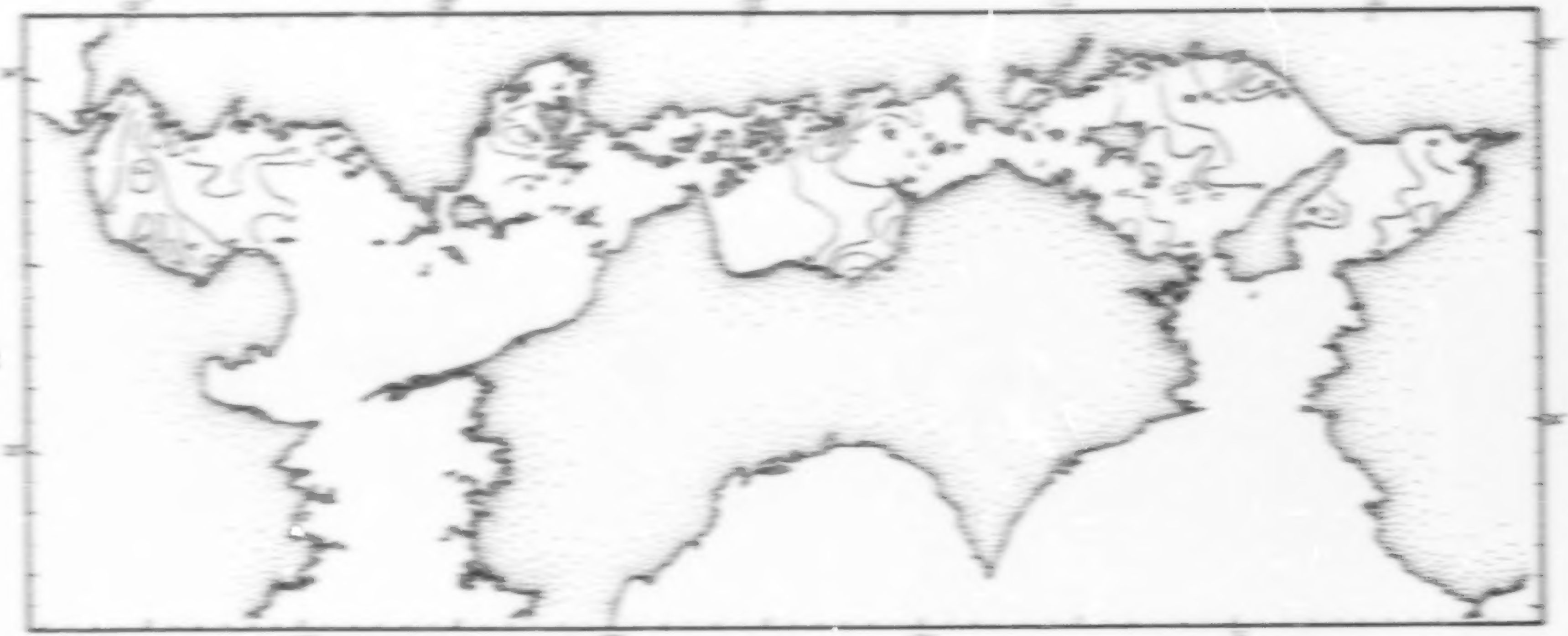


Figure 4. COD (mg/g) value of the bottom mud in the upper 5 cm layer in the Seto Inland Sea, Aug-Oct 1974.

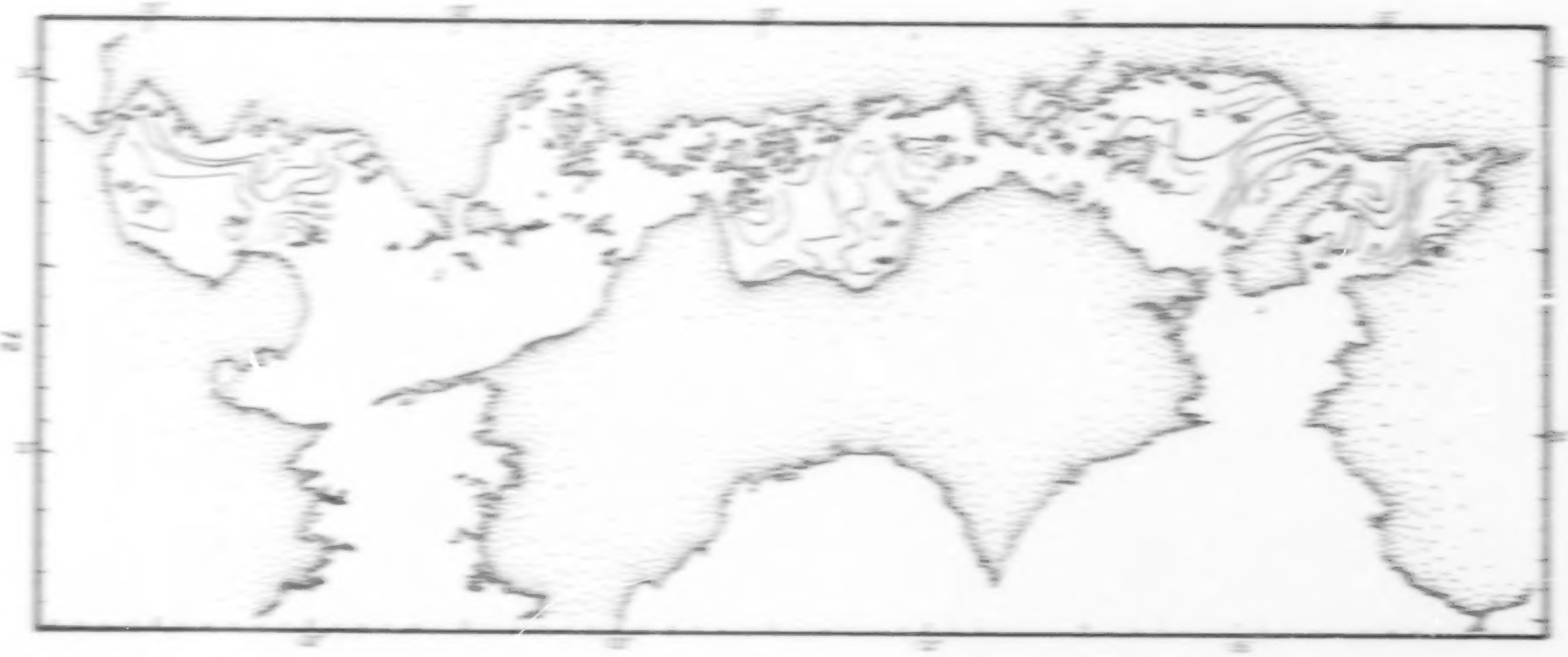


Figure 5. Ignition Loss (%) value of the bottom mud in the upper 5 cm layer in the Seto Inland Sea, Aug-Oct 1974.

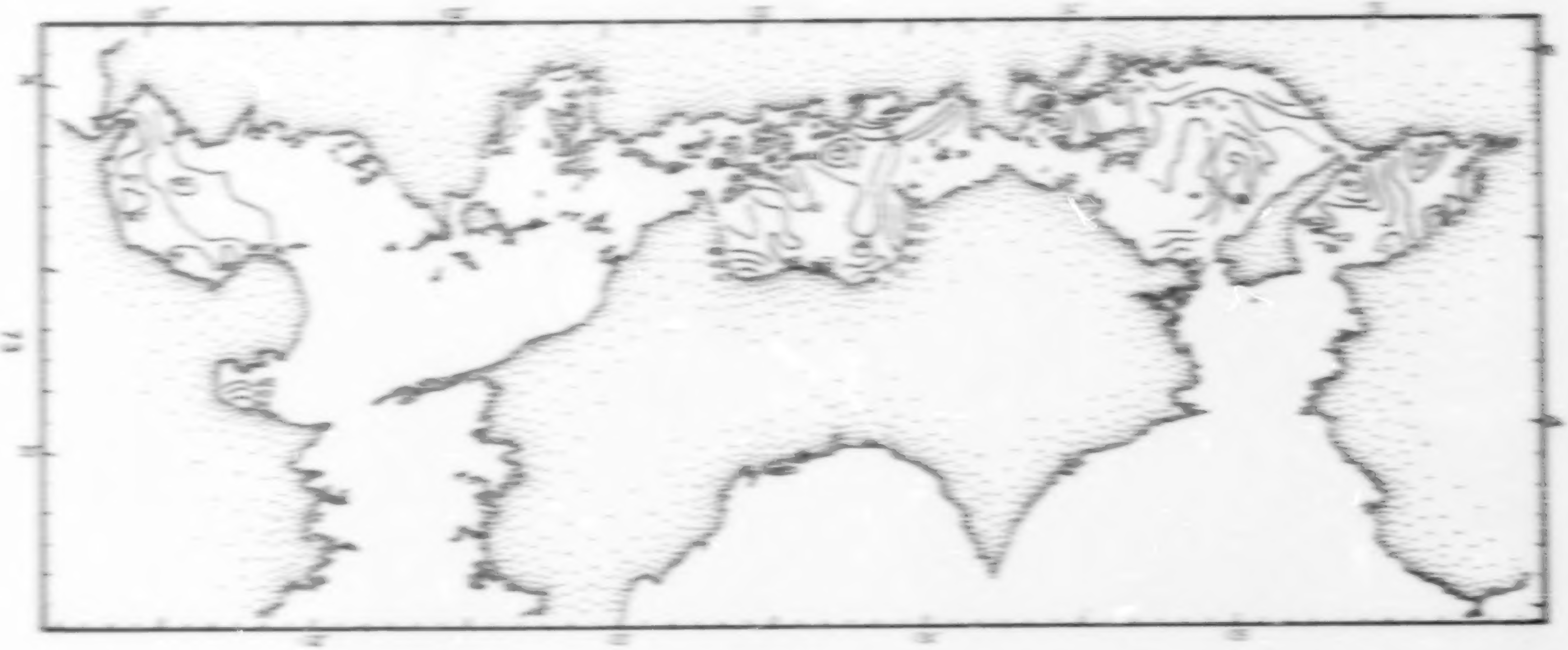


Figure 6. Total-S (mg/g) content of the bottom mud in the upper 5 cm layer in the Seto Inland Sea, Aug–Oct 1974.

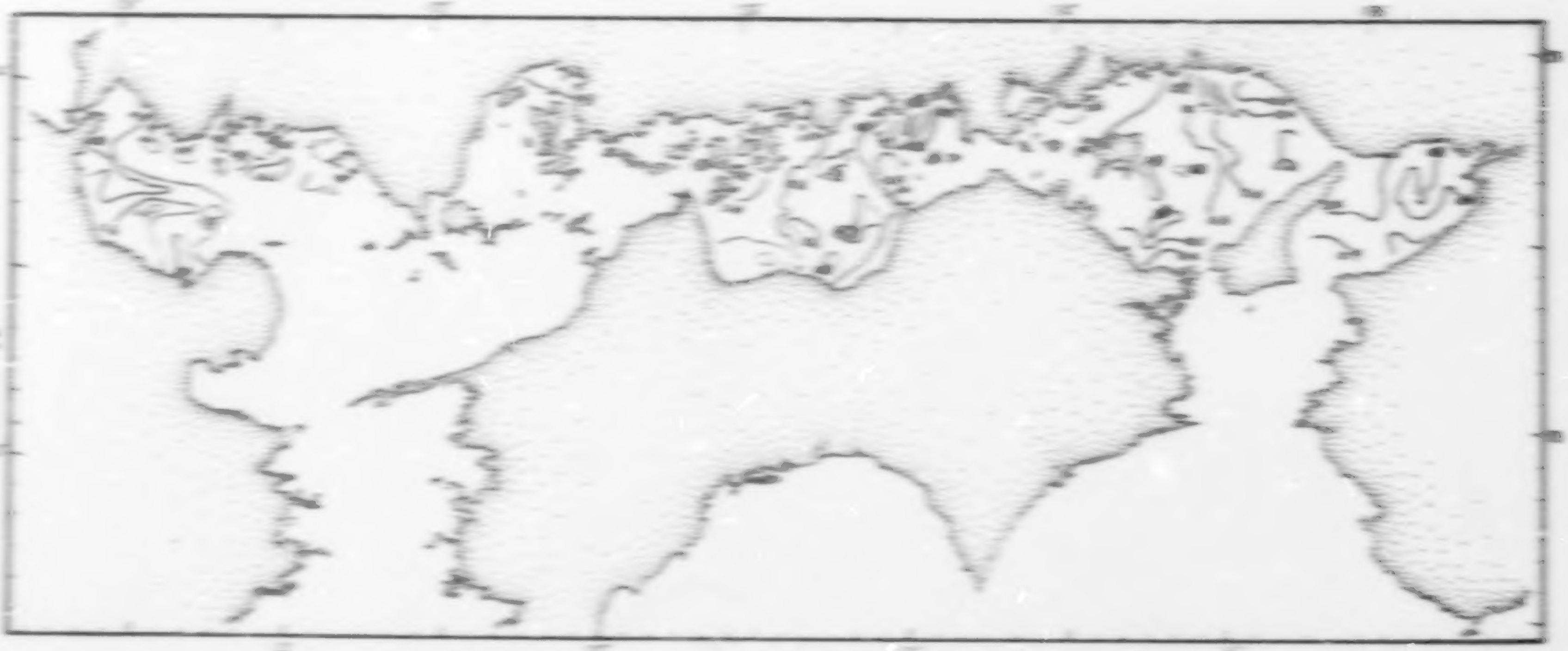


Figure 7. P (mg/g) content of the bottom mud in the upper 5 cm layer in the Seto Inland Sea, Aug–Oct 1974.

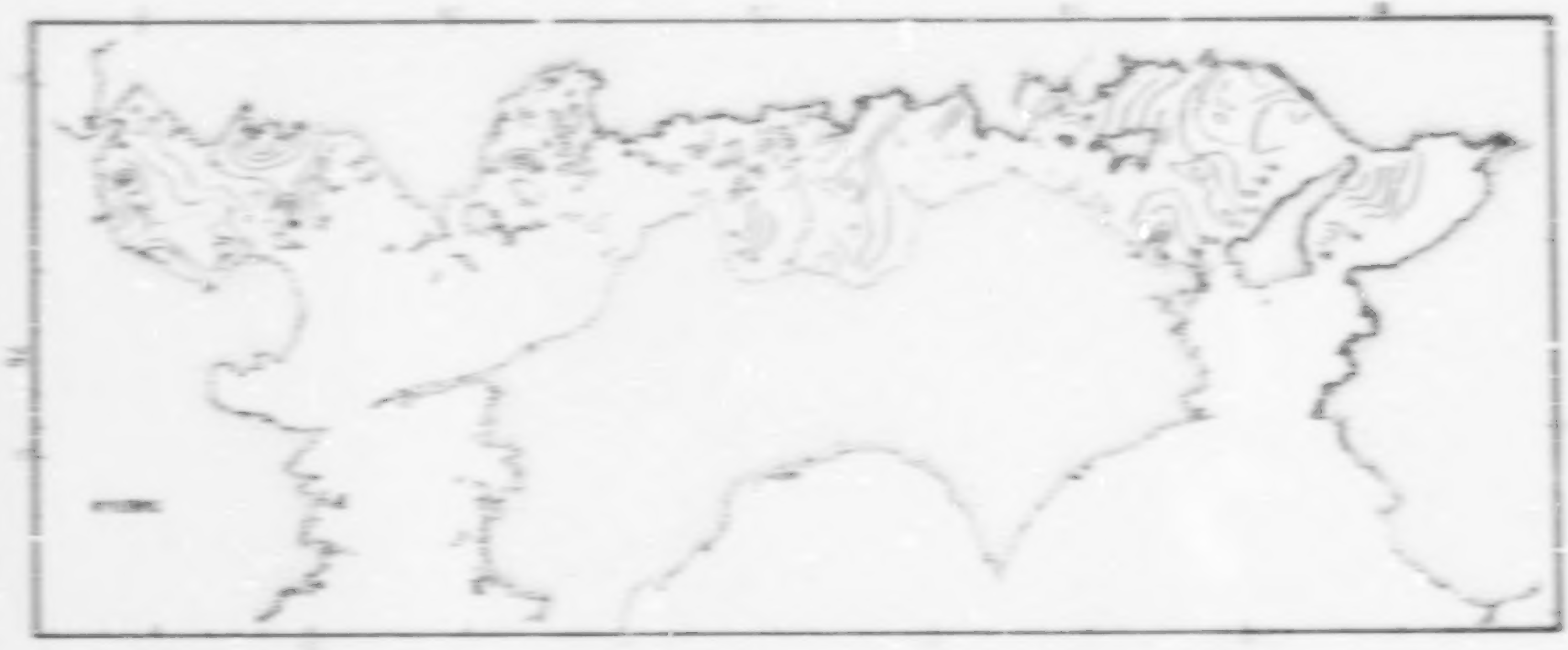


Figure 8. N (mg/g) content of the bottom mud in the upper 5 cm layer in the Seto Inland Sea, Aug-Oct 1978.

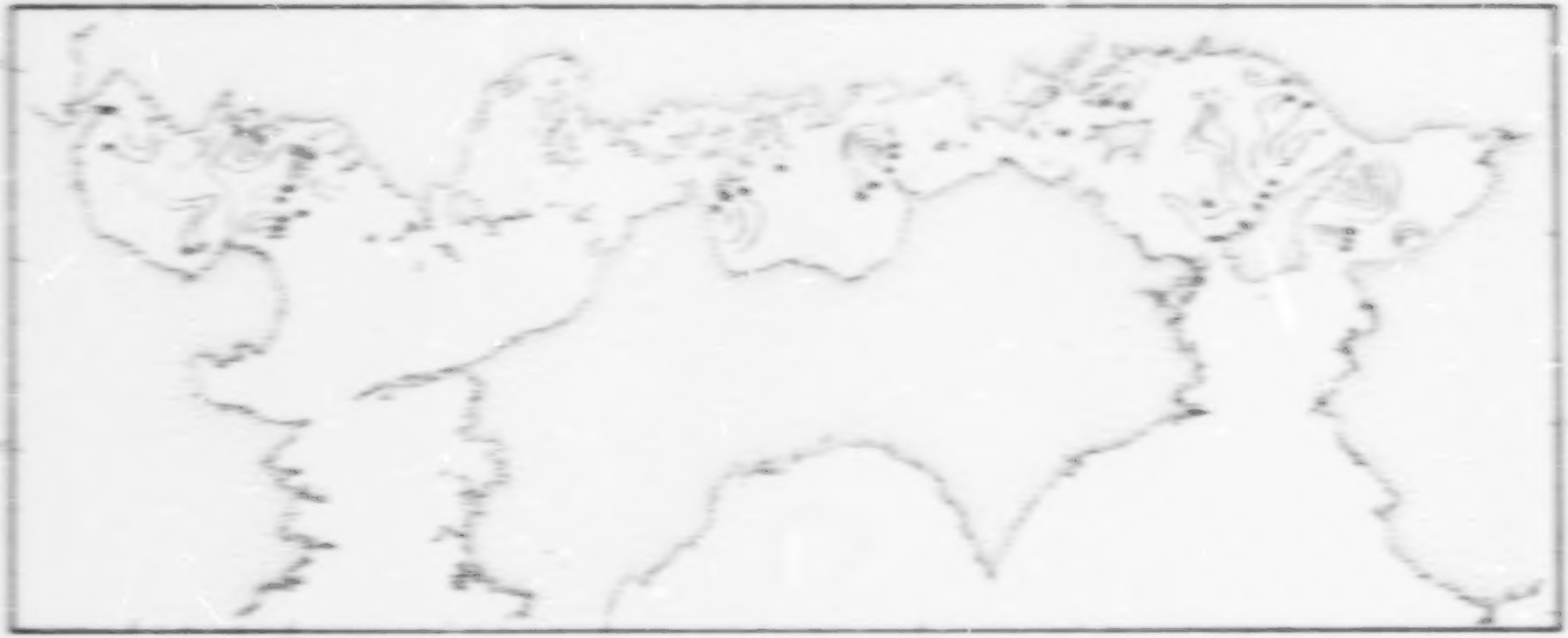


Figure 8. Composition (%) of silt (dia 125 μ) of the bottom mud in the upper 5 cm layer in the Seto Inland Sea, Aug–Oct 1978.

waste water from breweries. They are respectively 15 mg/g and 2-3 mg/g in the upper 20 cm of the mud column. The N/P ratio is 6 or 7. Generally the black surface layer is richer in nutrients than the greenish-gray deeper layer. Nitrogen is 2-3 mg/g, phosphorus is about 1 mg/g, and the N/P ratio is 3-10 in the surface layer. The N/P ratio is about 2.5 in the lower layer. Although nitrogen and phosphorus contents were not determined for the lower layer in the investigation in 1974 it usually has a nitrogen content of 1-2 mg/g and a phosphorus content of 0.5-1 mg/g.

The highest N/P (3.3) values were observed in Osaka Bay and Hiroshima Bay. Nitrogen content was 2.8 mg/g in both regions. On the contrary, the lower N/P ratios (1.1 and 1.6) were obtained in Iyo Nada and Kii channel respectively, and the nitrogen content was 1.2 mg/g in Iyo Nada and 1.3 mg/g in Kii channel.

In 28 regions the coastal areas were more polluted than the offshore areas. As shown in Table 4 the heavily polluted areas were off Otake and Iwakuni in Hiroshima Bay, Hiro Bay in Aki Nada, the inner part of Beppu Bay, off Mishima and Kawanoe in Hiuchi Nada and off Sakai in Osaka Bay. The major pollution sources are paper mill waste water in Hiro Bay, off Otake and Iwakuni and off Mishima and Kawanoe, and brewery waste water off Hofu, sewage off Sakai, and sewage and ironwork and oil refinery waste waters at the inner part of Beppu Bay. The characteristics of the sediment pollution are related to the kind of pollutant. For example, in the area polluted by the paper mill outfall COD and IL values are high. Brewery waste water causes high nitrogen content but low values of COD and total-S. Total-S and nutrient values are high in the area polluted by sewage.

Until 1973 about 30,000 kl of human waste were discharged per day in the Seto Inland Sea; 740 kl were released in the central part of Osaka Bay and 620 kl in Hiroshima Bay. Sewage released off Otake and Iwakuni in Hiroshima Bay caused high concentrations of nitrogen and phosphorus in this region. The distribution of COD in the upper 5 cm of the sediments at 71 stations off Iwakuni is shown in Figure 10. In Otake, 10^5 tons per day of paper mill waste water are released. In Iwakuni, 3 x 10^5 tons per day of paper mill waste water are released. Refineries, the petrochemical industry, the rayon factory and the airport waste water are other important sources of pollutants in these areas. Until the 1970's, untreated pulp mill waste water was discharged to the sea causing a brown colored bubbling water mass along the coast. Near the outfall, COD of the bottom mud was about 40 mg/g and remained over 30 mg/g as far as 3-4 km from the coast. The environmental water quality standard for fisheries (4) sets the desirable COD of the bottom mud at less than 20 mg/g. Sediments which met this quality were observed only around Atada Island as far as 4-5 km from the coast. The bottom mud off Iwakuni and Otake has been seriously polluted since the 1950's. In October 1961 COD was 40-56 mg/g off the coast between the Oze and Imazu Rivers.

The vertical distribution of COD varies throughout the Inland Sea. At the station having over 30 mg/g in COD in the upper 5 cm of bottom mud, the value is usually over 20 mg/g in the upper 30 cm. There is a station (Iwakuni St. 5) where COD is 31.8 mg/g in the upper 5 cm, 33.0 mg/g in the 15-20 cm layer, 21.3 mg/g in the 30-35 cm layer, 21.8 mg/g in the 81-86 cm layer, and

TABLE 4. QUALITY OF BOTTOM MATERIALS IN SPECIAL SELECTED SEA REGIONS IN THE SETO INLAND SEA, AUG. - SEPT., 1974

Item \ Sea Region		Hiro Bay	off Otake	off Iwakuni	off Hofu	off Mishima and Kawanoe	off Sakai
COD (mg/g)	Max.	55.7	51.4	47.1	36.5	46.8	23.7
	Av.	18.5	27.6	26.4	9.7	16.5	15.7
	Min.	10.1	12.6	15.9	3.7	5.9	7.6
IL (%)	Max.	13.2	27.8	16.4	19.6	19.1	14.4
	Av.	10.2	14.1	13.0	7.3	11.4	11.4
	Min.	6.2	7.5	10.7	2.2	3.5	7.9
Total-S (mg/g)	Max.	1.2	0.7	1.6	0.3	1.2	2.1
	Av.	0.2	0.2	0.4	0.1	0.2	1.0
	Min.	0.02	0.03	0.07	0.01	0.01	0.2
P (mg/g)	Max.	0.8	0.8	1.0	0.8	1.0	1.1
	Av.	0.6	0.7	0.7	0.5	0.7	0.7
	Min.	0.4	0.4	0.3	0.2	0.4	0.4
N (mg/g)	Max.	2.5	3.5	2.9	3.9	2.8	2.7
	Av.	1.8	2.5	2.2	1.1	1.7	2.1
	Min.	1.4	1.4	1.8	0.3	0.6	1.4
Composition of Mud (%)	Max.	99.6	99.1	99.9	99.4	100.0	100.0
	Av.	89.8	94.3	96.7	63.0	86.2	99.5
	Min.	38.6	38.4	72.9	2.5	3.3	96.0

mg/g in the 130-150 cm layer of the mud column. Off Kawanoe and Mishima the maximum COD is 46.8 mg/g in the upper 5 cm layer. This is due to paper mill waste water which is as great as that in the Iwakuni and Otake district.

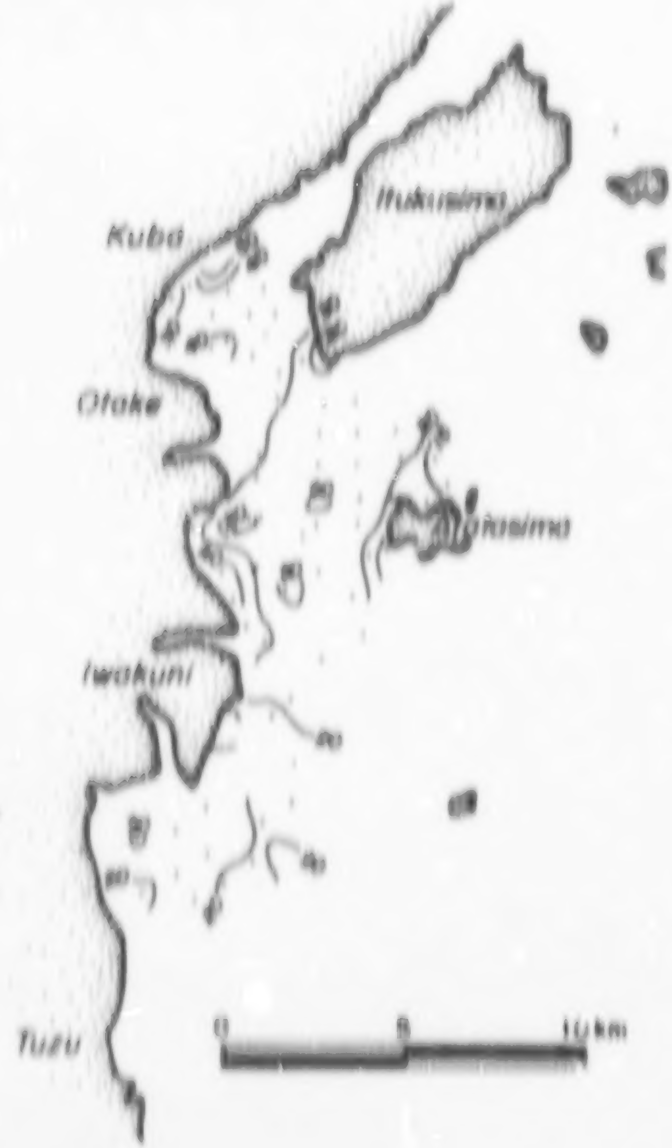

Itukusima
Kuba
Otake
kisima
Iwakuni
Tuzu
0
5
10 km

Figure 10. COD (mg/g) value of the bottom mud at upper 5 cm layer off Otake and Iwakuni, 23-25 August 1974.

COD in the upper layer of the bottom mud is not very different in the two sea regions, but the layer of high COD in the sediment is restricted only to the thin surface layer off Mishima and Kawanoe. The distriuution of polluted mud (COD over 20 mg/g) is not available in the deep layers over 25 cm in that region.

The pollution distribution is summarized as follows: the region off Otake and Iwakuni is located in the western part of Hiroshima Bay where water exchange is limited by the offshore current. The area off Mishima and Kawanoe is located at the eastern part of Hiuchi Nada and is not closed by the offshore current. Thus, the polluted mud is spread all over the eastern part of Hiuchi Nada. Paper mill waste water, 10^5 ton per day, is discharged into the inner part of Hiro Bay. In this region the area of polluted mud is restricted to a narrow area near the outfall. Distribution of high COD value stations is limited to a circle having the center at the point of the pollution source. Values over 30 mg/g lie as far away as 1 km from the drainage, and values of 20 mg/g are found as far away as 2 km. Vertically, COD is over 20 mg/g in the upper 50 cm of the bottom mud, and 30 mg/g in the upper 25 cm. Hiro Bay is narrow but has a wide mouth opening to Aki Nada. The exchange of water in and out of this bay is rather uniform, so the mass of colored waste water occasionally extends to the outer part of the bay. Under these circumstances the pulp fiber in the waste water does not settle thickly on the bottom. The horizontal and vertical distributions of the polluted bottom mud are ruled by the currents and topography of the sea floor.

The amount of polluted sediment was estimated as follows: COD in each 25 cm layer of the mud column was obtained at each station from the vertical distribution of COD using values for grain composition and echo sounder results. Stations with a bottom consisting of sand or rock were excluded. The area was divided into several blocks by grouping mud-bottom stations. The volume of each sediment layer in every block was obtained by multiplying the area by the thickness (up to 25 cm). A cumulative curve of the polluted sediment layer was drawn for each region examined (as shown in Figure 11) for the upper 25 cm layer in Hiroshima Bay. These curves are calculated with the percentage of the mud volume in every layer as vertical axis, and the average COD in each block as the horizontal axis. From this graph the volume of the polluted mud in each COD range can be estimated by multiplying the total volume of the mud by the percentage given in the cumulative curve. The amount of the polluted sediment estimated by COD at each region in the Seto Inland Sea is shown in Table 5. The volume of polluted sediment over 30 mg/g of COD is about 8 x 10^6 m^3 in the Seto Inland Sea, 92% of which is in Hiroshima Bay. For 40 mg/g there is a volume of 10^6 m^3 and 85% of it is found in Hiroshima Bay. Although pollution is slight in Aki Nada and Iyo Nada because of the relatively strong currents, a polluted bottom exists at Hiro Bay in Aki Nada and the inner part of Beppu Bay in Iyo Nada. This is shown in Table 5.

According to research done in August, 1975, the volumes of the polluted sediments in the bays of Ise and Mikawa which contain over 30 mg/g and 40 mg/g of COD are respectively 488 x 10^5 m^3 and 71 x 10^5 m^3. Although the area of Ise and Mikawa Bay is a tenth of the Seto Inland Sea, seven times the volume of polluted mud exists in Ise and Mikawa Bay. This is probably due to

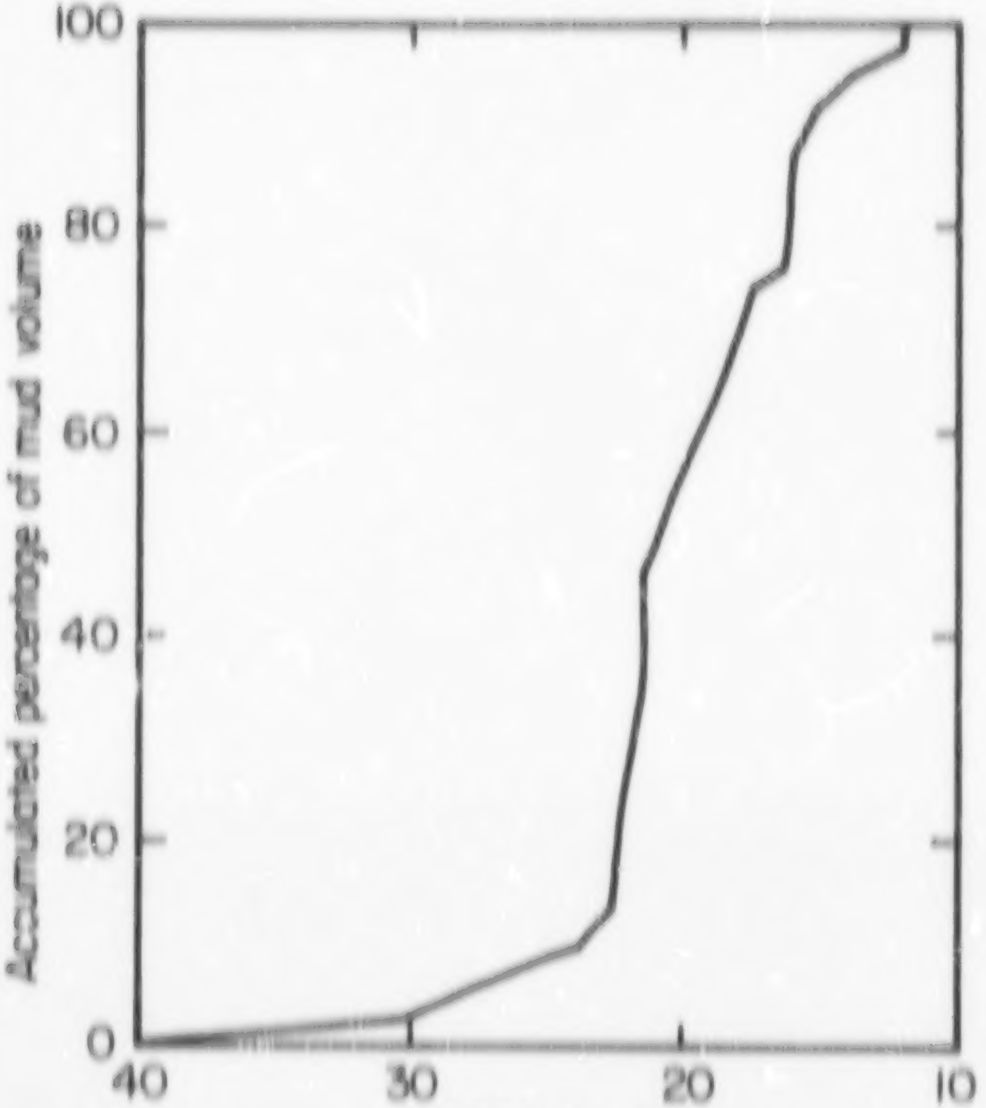


Figure 11. Cumulative curve of polluted sediment in the upper 25 cm layer in Hiroshima Bay, August 1974.

the difference in water exchange. According to the Environment Agency, the pollution load of COD was 1,600 tons per day in the Seto Inland Sea in 1972, and 600 tons per day in Ise and Mikawa Bay in 1973.

TABLE 5. VOLUME OF POLLUTED BOTTOM MUD IN SEVERAL SEA REGIONS IN THE SETO INLAND SEA, AUG. - OCT., 1974. X 10^4 M^3

Sea Region	Area (km^2)	COD mg/g 15	20	30	40	50
Osaka Bay	1,529	5,523.1	1,211.2	4.5	***	***
Kii Suido	1,554	21.3	***	***	***	***
Harima Nada	3,462	1,964.5	589.6	7.1	***	***
Bisan Strait	916	61.9	***	***	***	***
Bingo Nada	954	1,400.1	464.4	***	***	***
Hiuchi Nada	954	878.1	119.8	14.4	6.4	***
Aki Nada	963	608.5	58.3	26.5	8.8	4.1
Hiroshima Bay	946	75,737.0	22,116.1	725.6	87.0	0.7
Beppu Bay						
Iyo Nada	3,974	4,076.5	353.5	12.3	***	***
Suho Nada	3,100	17,633.2	794.0	***	***	***
Total	18,316	107,904.2	25,715.9	790.4	102.2	4.8

EXPERIMENTAL DREDGING OF ORGANIC POLLUTED SEDIMENT IN THE FISHING GROUNDS

Red tide broke out frequently in the Seto Inland Sea in the mid 70's, as shown in Figure 12. Recently, the red tide has been noted about 300 times a year in the Seto Inland Sea. The kind of red tide plankton has changed from diatoms to flagellates. To put it succinctly, the red tide in the Seto Inland Sea is becoming worse. Particularly the red tide caused by Hornellia (Chloromonadophyceae) is a large-scale outbreak during the warm season from July to September, and damage to cultured yellowtail (fish) is severe. The number of cases of red tide harmful to fisheries (shown in Figure 12) is not correlated with the scale of the damage. For example, in the summer of 1972 Hornellia red tide in Harima Nada and the western part of Kii Suido, produced a record breaking kill of cultured yellowtails valued at about 2 million U.S. dollars. It is clear that the deleterious effects of the red tide in the Seto Inland Sea owes much to eutrophication of inshore waters. Since 1973, the Fisheries Agency has been removing the polluted mud to reduce eutrophication in order to avoid fisheries damage.

The experiment was carried out as a joint venture of "World Ocean System Co." and "Fuyo Ocean Development Co." Dredging was done at Yura in Awaji Island in 1973, at Sagoshi Bay in Ako city in 1974, and at Aboshi Bay in Himeji city in 1975. In 1976 it will be carried out at Ie Island in Harima Nada. In this project the organically polluted sediment in the yellowtail rearing areas and off the river mouth is dredged to prevent diffusion of mud. The dredged mud is transferred to sedimentation pontoons, 600 m away, by a suction pipe on the sea surface. Then the mud is treated through the processes of coagulation, sedimentation and dehydration. The separated water is filtered, then discharged to sea. The settled sludge is hardened, then dumped on a reserved land area. The flow sheet of the experiment carried out for Aboshi Bay is shown in Figure 13. The bottom mud off the mouth of the Ibo river is black in color, smells of hydrogen sulfide, has 34-85 mg/g of COD and contains much sulfide and chromium.

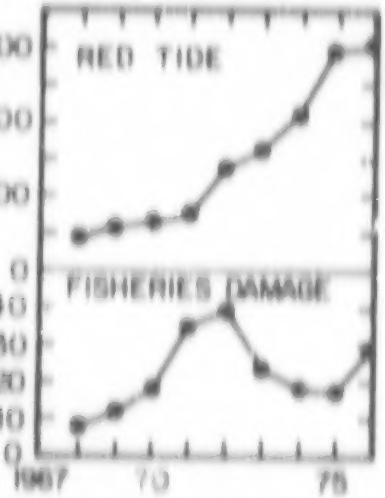


Figure 12. Frequency of red tide and fisheries damage in the Seto Inland Sea, 1967-1975

The bottom sediment dredge is equipped with a suction mouth which has movable wings and a variable volume (0-1500 m^3/hr) volute pump which dredges the polluted bottom mud at a speed of 1900 m^3 of slurry per hour. The mud to be dredged is 50-60 cm thick, 5700 m^2 in area and 2-2.5 m under the sea surface. The sediment is 30% in its average concentration of mud and has a D_{50} of 11 μ. The dredged mud is about 3100 m^3 in volume. Two sedimentation pontoons of 600 m^3 volume are used, one for sedimentation and the other for discharging the sludge. The volume of the sludge is 30 to 50% of the dredged mixture. Often the screen is clogged with deposited vinyl sheets or grass, so the volume and the mud concentration of the supernatant varies. The treatment pontoon has a capacity of 80 m^3 per hour. Alum and polymer are poured into the supernatant as a coagulant, the proper pH is regulated with sulfuric acid and sodium hydroxide, then sodium hypochloride is used for denitrification. The raw water is treated with polymer which settles out (dehydrater). Finally, the supernatant is treated with activated charcoal to remove the residual chlorine and the water is discharged to the sea after confirming its non-toxicity to marine organisms with a bioassay using minnows. The effluent water quality standard is described as follows: "SS <20 mg/l, COD <20 mg/l, NH_4-N <1 mg/l, free chlorine <0.05 mg/l, H_2S <1 mg/l, pH <9" and the heavy metals and harmful substances are discharged in concentrations less than the quality standard as given in the Water Pollution Control Law. The settled sludge in the sedimentation pontoon and the dehydrated cake from the dehydrater are mixed with cement, then dumped on the reclaimed land and the surface covered with sand.

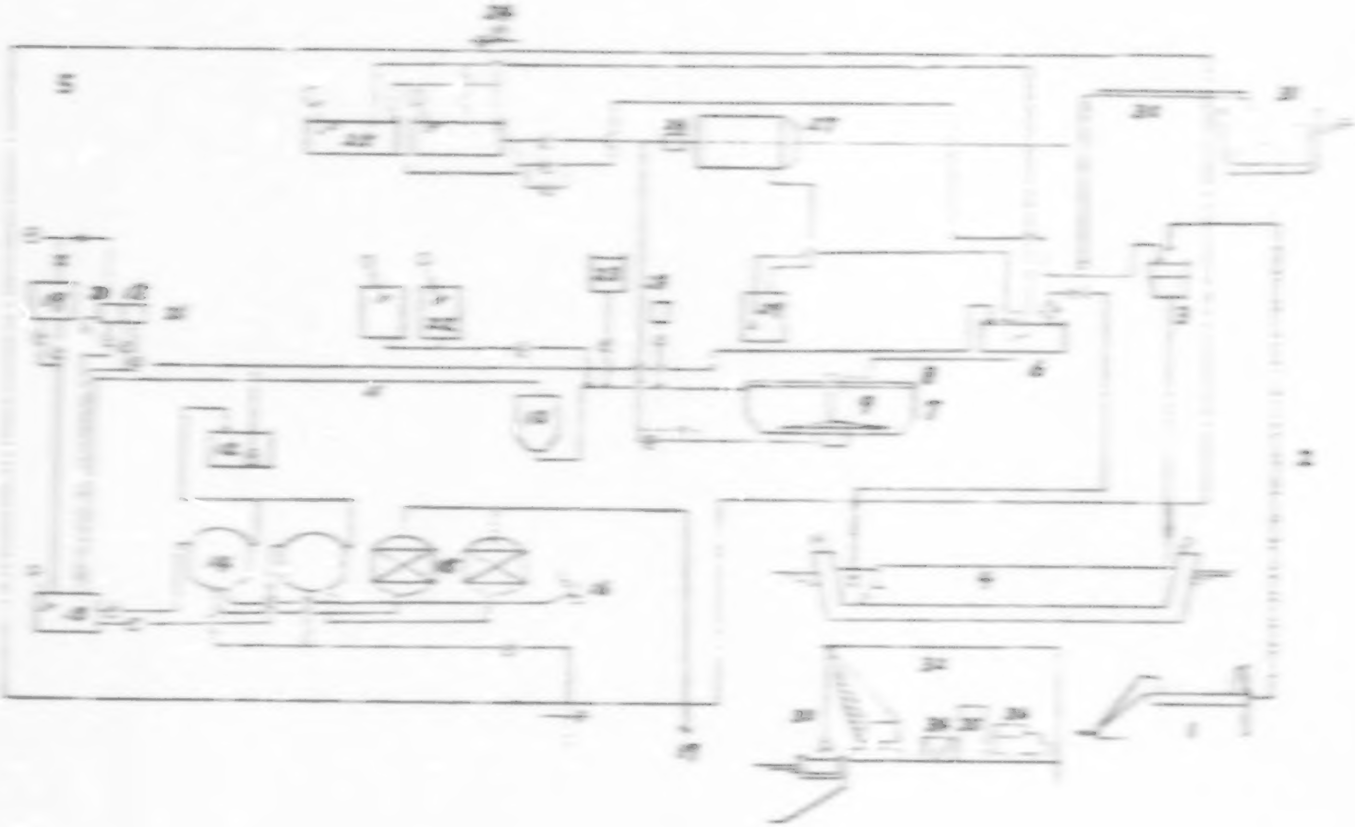

26
5
27
31
6
8
9
7
2
4
1

Figure 11. Flow sheet of withdrawal and treatment of polluted bottom mud.

(Fig. 13)

1 dredge (180 m^3/h)
2 pipe on sea surface
3 screen
4 storage barge
5 treatment pontoon

6 coagulation tank
7 sludge collector
8 concentration tank
9 thickener (140 m^3)
10 super-high rate pelletizing and sedimentation unit (pbs type)

11 separated water
12 back washing wastewater tank
13 chlorinated reaction tank
14 filter
15 activated carbon adsorption

16 back washing blower
17 effluent
18 sulfuric acid storage tank
19 sodium hypochloride storage tank
20 sodium hydroxide storage tank

21 polymer dissolving tank
22 P.B.S. polymer dissolving tank
23 aluminum sulfate storage tank
24 sea water pump
25 polymer dissolving tank

26 sludge mixing trough
27 dehydrater
28 sulfuric acid storage tank
29 separated water tank
30 belt conveyor

31 barge (300 m^3)
32 discharging wharf
33 clamshell bucket
34 hopper
35 cement silo
36 mixing car

Essentially this experiment is aimed at removing an organically polluted sediment without affecting the sea or increasing the red tide. Therefore, the effect on the surrounding sea and the red tide is estimated during the experiment. As a result there was no direct effect due to the experiment except the increase of NH_4-N concentration in sea water.

An experiment with red tide plankton culture was carried out to determine the effects of the effluent on the outbreak of the red tide. Cultures of *Hornellia* and *Olithodiscus* were added with sea water to the filtered effluent. They were kept for two weeks under the following conditions: water temperature, 20-30°C; illumination, 4,000 Lux for 12 hours per day. Neither culture increased in growth rate as a result. On the contrary, the growth rate was inhibited, and sometimes the *Hornellia* culture (and also minnows used to monitor the harmlessness of the effluent) were killed, probably due to the high concentration of NH_4-N and the residual chlorine and chloramine in the effluent.

On this project it was most difficult to determine how to deal with newly deposited materials on the surface of polluted mud. As mentioned above, the eutrophied bottom sediment acts as a secondary source of nitrogen and phosphorus for the bottom water. These nutrients are mainly released from the upper 15 or 20 cm of the sediment. But the suspended detritus in sea water is a more important secondary source. It settles gradually and concentrates in the water layer 1 or 2 cm above the bottom sediment, finally settling out as newly deposited material. So generally, in many polluted areas, there is a newly deposited material of a different color, several mm in thickness, which contains concentrated detritus from the water layer and is in contact with the bottom surface. It is the same situation as the case of fiber waste from pulp mills. In this project the mud was dredged by a dredging pump to prevent the diffusion of mud, but this is not always applicable to the newly deposited materials or the concentrated detritus. It is important to examine this problem, especially in the case of bottom sediment polluted by pulp fiber.

When selecting an area to dredge it is necessary to understand the impacts on fisheries and the significance of the mud in polluting the environment. When the project is carried out commercially, the balance between the scale of damages and the cost of removal and treatment must be examined. But, it is more important that the removal must be decided reasonably in view of the pollution mechanism. Once the dredging is decided upon, the quality, the range, the thickness of the mud and the time to remove it must be examined through research on the distribution of bottom mud and detritus.

Parameters indicating the amount of pollution such as COD, IL, N, P, total-S and size composition of mud must be given. Moreover, toxicity, heavy metals and oil pollution may also be necessary parameters depending upon the circumstances. These parameters must be measured vertically and horizontally in as much detail as possible, not only on the mud but on the surface deposits and the detritus, along with the rate of release. Insoluble nitrogen and phosphorus in the bottom mud do not accelerate eutrophication of the sea water, but once these nutrients change into the soluble state due to bacterial action or the lowering of pH in sea water, they are added to the bottom layer sea water through diffusion from the interstitial water in the bottom muds. Moreover, nitrogen and phosphorus also are released directly from the newly deposited materials and suspended detritus. These processes are very important in eutrophication of inshore waters. But, unfortunately, we have no effective method to sample the detritus, the new deposits and the interstitial water in mud, so it will be necessary to develop methods in the future. When the measurements are carried out in water which lacks dissolved oxygen, it is desirable to measure DO content continuously in time and space using a DO meter.

After bottom mud and the bottom water conditions are determined, then critical values for each parameter of the polluted mud must be determined to decide if removal is necessary. The desirable qualities of bottom sediment in fisheries grounds are given in environmental water quality standards such as "COD below 20 mg/g, total-S below 0.2 mg/g, the concentration of the n-hexene soluble matter less than 1% and no harmful substances in soluble state."(4) It is a matter of record that the qualities of bottom mud in the stagnant region of the Seto Inland Sea are much worse than those in the standard. Therefore, these values in the standard cannot be adopted as a lower limit of quality for removing bottom mud. In this experiment 30 mg/g of COD are decided as one of the criteria for removing organic polluted mud. But, according to the results presented from the economical point of view, the criteria might be considered "40 mg/g in COD, 15-20% in IL, 1 mg/g in total-S, 1 mg/g in total-P, 2-3 mg/g of total-N."

As mentioned above, the minimum quality of the polluted mud removed should be decided case by case from the view point of the pollution mechanism rather than the cost. The thickness of the mud to be removed may be 20 to 30 cm in general. The distribution of the polluted mud must be estimated vertically and horizontally by test bores in many stations and by investigation with an echo sounder. In conclusion, it is necessary to quickly establish standards for removing mud in order to carry out this project.

The project of removing polluted sediment should be completed with estimates of the improvement of the fisheries environments. This method has not been discussed yet in our research, but would be desirable to estimate the effects not only on improvement of the environment and reduction of the fisheries damage, but in technical problems such as the efficiency of removal.

CONCLUSIONS

In marine pollution, the surface water turbidity or colored waste water are directly recognized as problems. But toxicity and heavy metal contamination of the fisheries catch or bottom mud pollution cannot be as easily detected. They can be recognized the first time by a red tide or a polluted catch and the fish kills. For bottom sediment pollution in particular there are few directly obvious results. It is very difficult to carry out the research itself because the pollution and its damage occur at the bottom of the sea. But in the Seto Inland Sea the damages are becoming comparable to other pollution sources. Thus, bottom mud pollution and its means of influence are gradually becoming understood. Dredging bottom mud is now being recognized as an important countermeasure to marine pollution. However, the dredging and reclamation are frequently carried out in inshore waters, so the polluted bottom mud cannot be disregarded as a secondary source to add to the pollution load of the sea. It is natural that the regulation of the reclamation and the pollution load is a basic countermeasure to marine and bottom mud pollution. But in the polluted inshore waters such as the Seto Inland Sea, it is also important to remove the pollution load in the form of bottom mud. The progress of research on the polluted sediment is desirable principally for the protection of the marine environment.

REFERENCES (all in Japanese)

1) Report of the Basic Reseach on Improvement and Recovery in Fisheries Ground. (1974, 1975). Fisheries Agency, '75, '76.
2) Report of the Circulation of the Pollution Load in Ecosystem and the Index Species for the Pollution. (1973). Tokai and Nansei Reg. Fish. Res. Labs., Fish. Coll., '74.
3) M. Ukita, et al. On Some Problems in the Estimation of Released Nitrogen and Phosphorus from the Bottom Mud. J. Water and Waste, 17-10, 11, '75.
4) Water Quality Standard for Fisheries. Japan Fisheries Resources Conservation Assoc., '72.
5) T. Nitta, et al. Study on Pollution by Industrial Sewage. Bull. Naikai Reg. Fish. Res. Lab. 3, '53.
6) Report of the Realization Project of Preventing and Removing the Red Tide. (1974, 1975). Japan Fisheries Resources Conservation Assoc., '75, '76.

THE MECHANISM OF METHYLMERCURY ACCUMULATION IN FISH

M. Fujiki,[*] R. Hirota,[**] and S. Yamaguchi[*]

ABSTRACT

The factors contributing to methylmercury accumulation in the red sea bream (*Chrysophrys major*) were investigated by using sea water containing methylmercury, bottom sediment from Minamata Bay (methylmercury: 0.015 mg/kg [dry wt.], total mercury: 192 mg/kg [dry wt.]), and bait containing methylmercury. The contaminated sea water contained 0.5 µg/liter of methylmercury and fish placed in this water accumulated methylmercury in the body, going from a concentration of 0.012 µg/g (muscle tissue) to 0.033 µg/g (muscle tissue). The fish fed on methylmercury bait (0.133 µg/g) accumulated a little methylmercury; the methylmercury concentration in the fish increased from 0.012 µg/g (muscle tissue) to 0.020 µg/g (muscle tissue). Fish raised in a rearing tank containing bottom sediment from Minamata Bay did not show an effective accumulation of methylmercury; and methylmercury accumulation was almost the same as that of the control group.

INTRODUCTION

When Minamata disease was first recognized among inhabitants in the vicinity of Minamata Bay, the fish from Minamata Bay contained a high concentration of methylmercury. Since the discharge of waste water containing methylmercury from the factory was stopped, the methylmercury concentration in fish from Minamata Bay has gradually decreased. The methylmercury concentration in the fish from Minamata Bay has consequently reached about 0.4 mg/kg (wet weight). The concentration is not always lower than that in fish from the control area.

However, the mercury concentration in the bottom sediment of Minamata Bay is still very high. For example, the inorganic mercury concentration in the bottom sediment near Myojin-misaki is about 600 mg/kg (wet weight). It has been well known that methylmercury, formed from inorganic mercury, accumulates in fish more easily than does inorganic mercury. This is why extensive water pollution control is necessary in Minamata Bay.

---

[*] Department of Environmental Epidemiology, Institute of Community Medicine, the University of Tsukuba, Ibaraki, Japan

[**] Aitsu Marine Biological Station, Kumamoto University, Kumamoto, Japan

The mechanism of accumulation of methylmercury by fish is not completely known. However, it has been generally considered that methylmercury is taken up by the fish through the gills directly from sea water, from the digestive organs via food, or from the gills and/or digestive organs via bottom sediments. The following experiments were conducted to determine which of these speculations are correct and the degree to which each mechanism contributes to the problem.

EXPERIMENTAL METHOD

The red sea breams (Chrysophrys major) used in these experiments were obtained from a fish farm. Body length and weight of the fish is shown in Table 1. Two hundred of the red sea breams were divided into 4 groups, A to D, each group containing 50 fish. The red sea breams were acclimated to the conditions of the fish rearing tank (Figures 1 and 2) for 3 days before the experimental procedures.

Experiment A

Sea water containing methylmercury (0.5 μg/liter) was added to the fish rearing tank (1000 l) and aerated by an agitating pump. Then the sea breams were reared in the tank for 10 days. The sea water containing methylmercury was replaced each day. Ten of the sea breams were taken out of the tank every 2 days and the methylmercury in the fish was measured by gas chromatography.

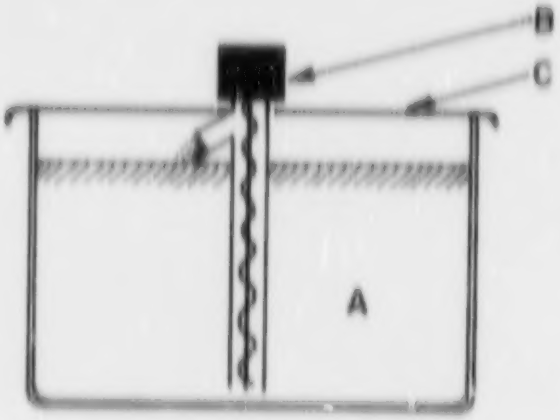


Figure 1. Experimental apparatus for
Experiment A, B, and D.
a: fish rearing tank (1000 l)
b: agitating pump
c: net

TABLE 1. THE MEAN OF THE BODY LENGTH AND WEIGHT OF THE RED SEA BREAM

Experimental Group	Rearing Time	0		2		4		6		8		10	
		L.	W.	L.	W.	L.	W.	L.	W.	L.	W.	L.	W.
A	Mean	12.2	42.6	10.7	33.7	11.5	38.2	11.0	37.5	11.3	38.8	11.9	41.2
	S.D.	1.3	12.4	1.4	11.1	1.1	12.6	0.9	8.7	0.7	7.9	0.8	5.9
B	Mean	12.2	42.6	11.3	39.4	11.2	40.4	11.3	40.0	11.3	37.6	11.8	41.9
	S.D.	1.3	12.4	1.2	9.5	1.0	8.6	1.4	14.6	0.8	8.5	1.0	9.6
C	Mean	12.2	42.6	11.8	40.5	11.7	41.3	11.7	43.7	11.3	37.9	12.0	42.3
	S.D.	1.3	12.4	1.8	14.0	1.1	9.1	1.4	10.2	0.8	8.1	1.0	6.2
D	Mean	12.2	42.6	11.1	32.9	10.9	35.0	11.1	38.3	11.5	40.0	11.5	35.9
	S.D.	1.3	12.4	1.2	8.3	1.0	9.5	1.2	10.0	1.0	9.2	1.2	7.4

Rearing Time: day Length: cm Weight: g

A: Reared in sea water containing 0.5 μg/l of methylmercury
B: Fed the prawn containing 0.133 μg/l of methylmercury
C: Reared in sea water containing the suspended solids and the bottom sediment from Minamata Bay
D: Control

Experiment B

Sea breams were reared in a tank (1000 l) holding aerated sea water for 10 days. Sea water was replaced daily. The prawn (Penaeus japonicus) was obtained from a fish farm and reared in the sea water containing methylmercury. Then the prawn was used as bait. The content of methylmercury in the prawn was 0.133 μg/g. The amount of the bait eaten was about 7% of the sea bream's body weight per day. Ten of the sea breams were removed from the tank every 2 days and the methylmercury in the fish was measured by gas chromatography.

Experiment C

Three kg of the bottom sediment from Minamata Bay containing 0.015 mg/kg (wet weight) of methylmercury and 192 mg/kg wet weight) of total mercury was placed on the bottom of the fish rearing tank (1000 l). Sea water was added to the tank and aerated by an agitating pump, and then the sea breams were reared in the tank for 10 days. The bottom sediment and sea water were replaced daily. Ten of the sea breams were taken out of the tank every 2 days and the methylmercury in the fish was measured by gas chromatography.

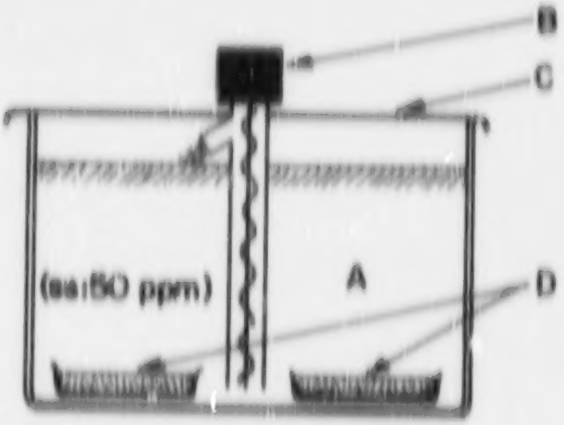


Figure 2. Experimental apparatus for Experiment C.
a: fish rearing tank (1000 l)
b: agitating pump
c: net
d: bottom sediment vessel

Experiment D

Sea water was added to the fish rearing tank (1000 l) and aerated by an agitating pump. Then the sea breams were reared in the tank for 10 days. Sea water was replaced daily. Ten of the sea breams were removed from the tank every 2 days and the methylmercury in the fish measured by gas chromatography.

The prawn (0.018 µg/g of methylmercury) obtained from a fish farm was used as the bait for each group except the B group. The amount of the bait eaten was about 7% of the sea bream's body weight per day. Each sample consisting of 10 of the sea breams was homogenized and then used to measure the methylmercury content. The temperature of the sea water during the 10-day experimental period was between 25.1 and 28.5°C.

RESULTS

The concentration of methylmercury accumulated in the sea bream is shown in Table 2 and Figure 3. The methylmercury concentration in the fish of the A group increased gradually from 0.12 µg/g (muscle tissue) to 0.033 µg/g (muscle tissue) for 10 days, that of the B group increased gradually from 0.012 µg/g (muscle tissue) to 0.020 µg/g (muscle tissue) for 10 days, that of the C group increased gradually from 0.012 µg/g (muscle tissue) to 0.016 µg/g (muscle tissue) for 10 days, and that of the D group increased gradually from 0.012 µg/g (muscle tissue) to 0.015 µg/g (muscle tissue) for 10 days. The methylmercury accumulation in the fish of the A group was evidently higher than that of the other groups. The methylmercury accumulations in the fish of the B and C groups were almost the same as that of the D group. However, the methylmercury accumulation in the fish of the B group was slightly higher than that of the C group and that of the C group was almost the same as that of the D group (the control).

TABLE 2. THE METHYLMERCURY CONCENTRATION IN THE RED SEA BREAM

Experimental Group \ Rearing Time	0	2	4	6	8	10
A	0.012	0.019	0.019	0.022	0.038	0.033
B	0.012	0.017	0.012	0.018	0.023	0.020
C	0.012	0.011	0.016	0.013	0.019	0.016
D	0.012	0.011	0.015	0.014	0.023	0.015

Rearing Time: day   methylmercury concentration: µg/g (wet wt. of muscle)

A: Reared in sea water containing 0.5 µg/l of methylmercury
B: Fed the prawn containing 0.133 µg/g of methylmercury
C: Reared in sea water containing the suspended solid and the bottom sediment from Minamata Bay
D: Control

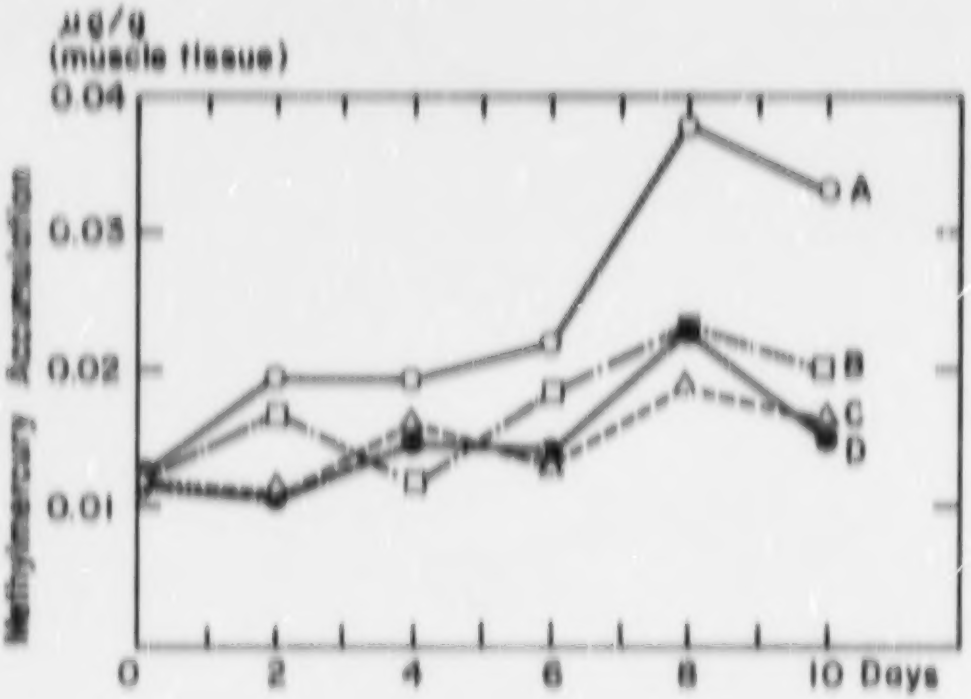


Figure 3. The relation between rearing time and methylmercury accumulation.
A: reared in sea water containing 0.5 μg/l of methylmercury
B: fed the prawn containing 0.133 μg/l of methyl mercury
C: reared in sea water containing suspended solid and bottom sediment from Minamata Bay
D: control

DISCUSSION

In experiments on methylmercury accumulation in fish, it would be more desirable to try a rearing experiment for 50-100 days or more. A special laboratory was not available to continuously supply the large amount of sea water required to rear marine fish. Conventional equipment was available only for this experiment so the term of the experiment was limited to 10 days.

Experiment A was designed to observe the accumulation of methlymercury from sea water via fish gills. Experiment B was designed to observe the effect of the methylmercury accumulation by the food chain. And experiment C was carried out to observe the effect on methylmercury accumulation by the intake of suspended solids containing methylmercury.

In experiment A the methylmercury concentration in the sea water was set at the concentration given as the maximum tolerance limits in Japan for water in the environment (0.5 μg/liter). The methylmercury concentration in the prawn used for experiment B was about 10 times as much as that in the prawn used for experiments A, C and D. Superfluous bottom sediment was used in experiment C because it was considered that both suspended solids containing methylmercury and dissolvable methylmercury from bottom sediments were cumulative factors.

Experiment D was the control group and the increase of methylmercury in the fish of experiment D has the same meaning as the increase occurring in the natural environment via the normal food chain. In experiment B, the methylmercury concentration of the prawn used as the bait was 10 times as much as that of the prawn in the natural environment. However, the methylmercury accumulation in the fish of the B group showed only a small increase. Therefore, it is concluded that the food chain is not necessarily an important factor for the accumulation of a large amount of methylmercury.

In experiment C, the concentration of suspended solids (SS) was 50 ppm in sea water and the superfluous bottom sediment was on the bottom of the tank. This condition is almost the same as conditions which occur during dredging work. The methylmercury accumulation in the fish of the C group was the same as that of the D group. Therefore, it is concluded that the suspended solids and the bottom sediments are not effective pathways for the accumulation of methylmercury in fish. In experiment A, the concentration of accumulated methylmercury in fish increased distinctly in spite of the low concentration of methylmercury in sea water (0.5 μg/liter). Therefore, it is concluded that dissolved methylmercury in sea water is one of the critical factors for methylmercury accumulation in fish.

CONCLUSION

It was found by these experiments on environmental methylmercury accumulation in marine fish that mercury in the suspended solids and bottom sediments did not accumulate in the fish, that the accumulation via the food chain was unexpectedly low, and that dissolved methylmercury in sea water was the critical agent for methylmercury accumulation.

DETERMINATION OF TRACE AMOUNTS OF METHYLMERCURY IN SEA WATER

H. Egawa and S. Tajima*
Department of Industrial Chemistry
Faculty of Engineering
Kumamoto University

ABSTRACT

In determining methylmercury (MeHg) concentration, gas chromatography after extraction with benzene and glutathione is common, but the concentration of MeHg in sea water is generally so low that this common method can not be directly used. In this study chelating resins which have a selective adsorption for MeHg were investigated as a means of preconcentrating MeHg in sea water.

The trace amounts of MeHg in sea water can accurately be determined in the concentration range from 0.005 μg/l to 0.05 μg/l (as Hg) after preconcentration of MeHg by adsorption and elution using chelating resin in a batch method.

Macroreticular chelating resin (A) containing episulfide groups was excellent for both the adsorption of MeHg and the elution of it with hydrochloric acid. The adsorption of MeHg on the resin (A) attained equilibrium after shaking for 12 hours at 30°C. The calibration curve of MeHg added to deionized water, synthetic sea water and natural sea water had excellent linearity in the concentration range of 0.005 μg/l to 0.05 μg/l and it was only slightly affected by mercuric ions and other various ions in sea water.

The following method is recommended. A 0.2 g chelating resin (A) is shaken with 1 liter of sea water for 15 hours at 30°C. MeHg in the elutriate is first extracted with benzene and then with glutathione solution from benzene. MeHg in glutathione solution is then analyzed by the flameless atomic adsorption technique. The concentration of MeHg is determined from the calibration curve.

---

*2-39-1 Kurokami, Kumamoto-shi, Kumamoto 860, Japan

This method was applied for the determination of MeHg in sea water samples collected in Minamata Bay and the adjacent sea.

INTRODUCTION

In the process of dredging bottom sediments containing mercury compounds the mercury compound may dissolve in sea water under certain conditions. The mercury content in sea water, particularly methylmercury (MeHg), must always be checked during the process to prevent secondary pollution. This is because the dissolved MeHg is accumulated directly by fish (1).

The concentration of MeHg in sea water is generally so low that the usual method cannot be applied directly to such a sample. The determination of trace amounts of total mercury, using flameless atomic absorption spectrophotometry after preconcentration of samples, has been reported by many authors. Nishimura et al. (2) reported that as low as 0.005 μg/l of total mercury in sea water, which is acidified with H_2SO_4 to stabilize the concentration of mercury, can be determined after preconcentration of mercury on silver metal particles. Baltisberger et al. (3) investigated the separation and identification of nanogram quantities of MeHg, Hg(I), and Hg(II) in an aqueous solution by using isothiocyanatopentaaquochromium in an ion exchange procedure. But they have not tested less than 0.1 μg/l of mercury in environmental water samples because of the problem of background contamination from reagents.

This paper investigates the method of determining trace amounts of MeHg in sea water by preconcentration, using chelating resins having selective adsorption for MeHg.

EXPERIMENTAL METHODS

Chelating Resin

The macroreticular chelating resins used in this study were prepared in our laboratory. They have selective adsorption for MeHg. The estimated structure of a functional group in these chelating resins is shown in Figure 1. For comparison the commercial chelating resin Diaion CR-10, containing amino diacetic acid groups, was also used.

Reagent

100 mg/l standard stock solution of mercury was prepared from special grade chemicals of methylmercurychloride and mercuricchloride obtained from the Wako Pure Chemical Industry LTD. 10 μg/l and 100 μg/l standard solutions were prepared before use from a 1 mg/l intermediate standard solution. The other reagents (hydrochloric acid, benzene, reduced glutathione etc.) used in this study were obtained from Wako Pure Chemical Industry Ltd. A synthetic sea water was prepared by the method of Lyman-Fleming (4).

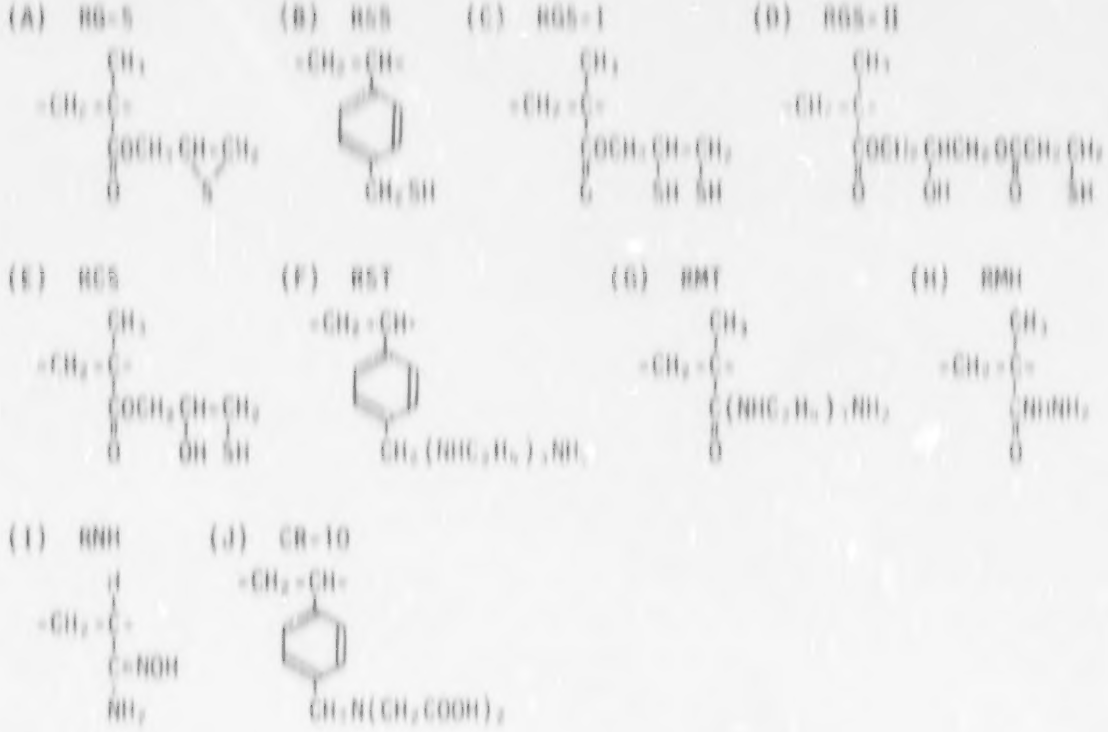


Figure 1. Macroreticular chelating resins.

Adsorption Procedure

A precise amount of chelating resin was placed in a stoppered Erlenmeyer flask, the aqueous solution containing MeHg was added to the flask, and the flask was mechanically shaken in a constant temperature bath at 30°C for the designated time (batch method). After the adsorption the resin was separated from the solution by filtration and rinsed with an adequate volume of deionized water and dried for 2 days at room temperature.

Elution Procedure

The resin was placed in a stoppered 100 ml Erlenmeyer flask, hydrochloric acid solution was added to the flask, and the flask was mechanically shaken in a constant temperature bath at 30°C for 3 hours. After the elution the resin was separated from the elutriate.

Extraction Procedure

MeHg in the elutriate was extracted to 100 ml of benzene after the concentration of hydrochloric acid in the elutriate was adjusted to one Normal, and then it was extracted with 10 ml of 0.05% reduced glutathione solution.

Analysis of mercury

The concentration of mercury in samples prepared in the above procedure was analyzed by the flameless atomic absorption technique, using the instrument of Rigaku Mercury SP (Rigaku Electric Company, Japan). The detection range of mercury used in this study was usually in the 0 to 5 ng range.

RESULTS AND DISCUSSION

Adsorption of Methylmercury on Chelating Resin and Subsequent Elution of It from the Resins

The chelating resins used in this study were synthesized in our laboratory and had selective adsorption for mercury in mole order concentrations. The first thing done was to examine the selective adsorptive ability of these resins for MeHg in low concentrations and the subsequent elution of MeHg with hydrochloric acid from the resins.

The following procedure was carried out. A 0.1 g chelating resin was placed in a stoppered 100 ml Erlenmeyer flask, 50 ml of 10 μg/l MeHg solution were added to the flask, and the flask was shaken mechanically for 3 hours at 30°C. The amounts of MeHg adsorbed on the respective resins were determined from differences in the initial and residual concentrations of MeHg. The resins separated after the adsorption of MeHg were placed in a stoppered 100 ml Erlenmeyer flask, 50 ml of 2N-HCl or 25 ml of 4N-HCl were added to the flask, and the flask was shaken mechanically for 3 hours at 30°C. MeHg in the elutriate was analyzed after the extraction procedure as described in the experimental section. The results are presented in Table 1.

The resins (A), (B), and (E) had excellent adsorption for MeHg. MeHg could be eluted completely from resins (A) and (E) by 4N-HCl but could not be eluted completely from resins (B) and (C). 4N-HCl was a better eluent of MeHg than 2N-HCl. MeHg was not decomposed by the acid. It was concluded from the above that the chelating resin (A), which contains episulfide groups, is the most adequate for the purpose of these studies.

Effect of Adsorption Time on Recovery of Methylmercury

The time taken for the adsorption of MeHg on the resin to reach equilibrium was investigated. 0.2 g of chelating resin (A) was placed in a stoppered 2 liter Erlenmeyer flask, 1 liter of 0.05 μg/l MeHg solution was added to the flask and the flask was shaken mechanically for periods ranging from 3 to 48 hours at 30°C. The elution was carried out under constant conditions. Because of the true concentration of MeHg used in this investigation, the MeHg adsorbed on the resin could not be determined from the residual concentration of MeHg in the solution after adsorption. Therefore, the recovery of MeHg through the procedure of adsorption, elution and extraction for each adsorption time was determined. The results are shown in Figure 2. It was found that adsorption equilibrium is reached after 12 hours of shaking at 30°C.

TABLE 1. ADSORPTION AND ELUTION OF METHYLMERCURY BY USING CHELATING RESINS

Resin symbol	Adsorption (1)		Eluent (3)	Elution (2)	
	MeHg adsorbed (μg as Hg/g-R)	Percentage of adsorption (%)		MeHg eluted (μg as Hg/g-R)	Percentage of elution (%)
(A) RG-5	4.37	95.0	α	3.80	87.0
	4.37	95.0	β	4.38	100.2
(B) RSS	4.53	98.6	α	1.99	43.9
	4.60	100	β	1.99	43.3
(C) RGS-I	4.30	93.5	α	3.11	72.3
	4.37	94.9	β	3.96	90.6
(D) RGS-II	4.13	89.9	α	3.43	83.1
	4.13	89.9	β	3.54	85.7
(E) RCS	4.30	93.5	α	3.85	89.5
	4.30	93.5	β	4.31	100.2
(F) RST	3.40	73.9	α	2.05	60.3
	3.36	72.8	β	2.83	84.5
(G) RMT	2.03	47.3	β	1.92	94.2
(H) RMH	3.40	73.9	α	2.95	86.8
	3.47	75.4	β	3.10	89.3
(I) RNH	2.17	47.1	α	1.77	81.6
	2.08	45.3	β	1.83	88.0
(J) CR-10	3.42	79.5	β	2.94	86.0

(1) 0.1 g of resin was shaken with 50 ml of 10 μg/l (as Hg) MeHg solution for 3 hours at 30°C. (2) Resin adsorbed MeHg was shaken with HCl for 3 hours at 30°C. (3) α: 2N-HCl ml, β: 4N-HCl 25 ml

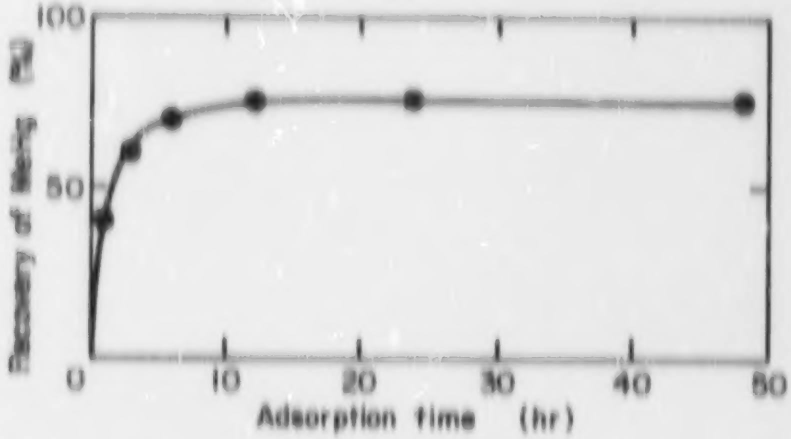


Figure 2. Effect of adsorption time on recovery of methylmercury. Adsorption: Resin 0.2 g/MeHg solution 0.5 μg/l (as Hg), shaking at 30°C.
Elution: 4N-HCl 25 ml, shaking for 3 hr at 30°C

The effect of the time interval between adsorption and elution on the recovery of MeHg was also studied. The results are shown in Figure 3. When the elution was carried out soon after the adsorption, the recovery of MeHg was slightly greater than in the case where the elution was done after 2 days. After that, the recovery of MeHg was not affected, even if the resin was allowed to stand at room temperature for over 2 days after the adsorption of MeHg.

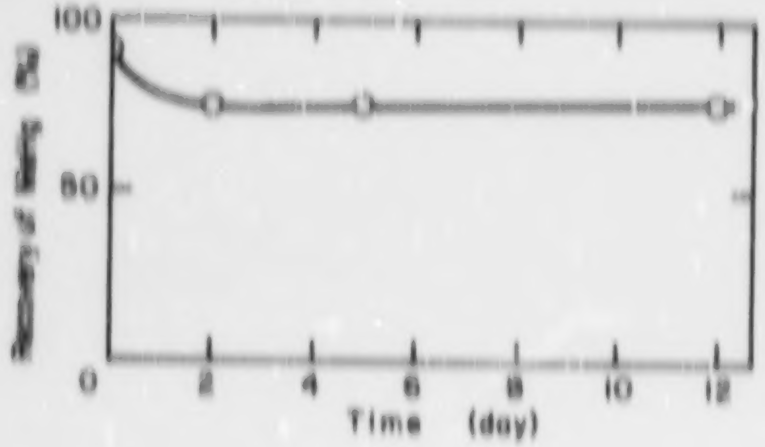


Figure 3. Effect of time interval between adsorption and elution on the recovery of methylmercury. Adsorption and elution: same as Figure 2.

This implies that trace amounts of MeHg in natural waters are able to be determined by preconcentration using chelating resins because the recovery of MeHg will be constant after the adsorption has attained equilibrium. Recovery will not be 100 percent, however.

Calibration Curve for Trace Amounts of Methylmercury

The calibration curve for the method under study was determined as follows: A standardized 10 μg/l MeHg solution was diluted with deionized water to obtain a series of MeHg sample solutions ranging in concentration from 0.01 μg/l to 0.05 μg/l (as Hg). The glassware was washed with an aqueous solution of EDTA or $Na_2S_2O_3$ and with a dilute acid solution and then rinsed with deionized water before use. This was to decrease blank values in this procedure. Then 0.2 g of chelating resin (A) were shaken mechanically with 1 liter of the sample in stoppered 2 liter Erlenmeyer flasks for 15 hours at 30°C. The resin was dried for 2 days at room temperature (after the separation from samples) and was then shaken mechanically with 25 ml of 4N-HCl in a stoppered 100 ml Erlenmeyer flask for 3 hours at 30°C. MeHg in the elutriate was extracted at first with benzene and then with glutathione solution from benzene. Through this entire procedure the trace amount of MeHg in 1 liter of sample was finally concentrated in 10 ml of 0.05% glutathione solution.

One ml of the glutathione solution was analyzed by the flameless atomic absorption technique. The data represented the MeHg recovered (reported as ng of Hg.

The results shown in Figure 4 illustrate that the calibration curve has an excellent linearity for concentration of MeHg. It has been found that MeHg concentration in water samples can be determined from the calibration curve. It is considered that the datum for 0 μg/l (no addition of MeHg) is attributed to the reagents used in this procedure.

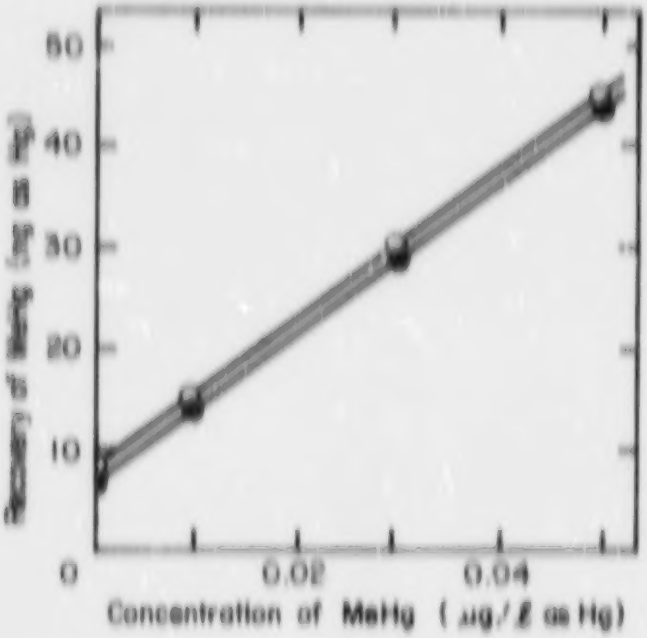


Figure 4. Calibration curve of methylmercury added to deionized water.
●First time
○Second time

Effect of Inorganic Mercury on the Calibration Curve of Methylmercury

Inorganic mercury compounds (Inorg Hg) usually exist in greater quantity than MeHg in natural water samples and are also adsorbed on the chelating resins. The effect of Inorg Hg on the MeHg calibration curve was investigated.

A calibration curve of MeHg in the concentration range of 0.01 μg/l to 0.05 μg/l plus Inorg Hg, was determined by the procedures outlined in the previous section. The concentration of Inorg Hg in the solution used in this experiment was 0.5 μg/l as mercury. This value was based on the limiting value in the natural environment. The results are shown in Figure 5.

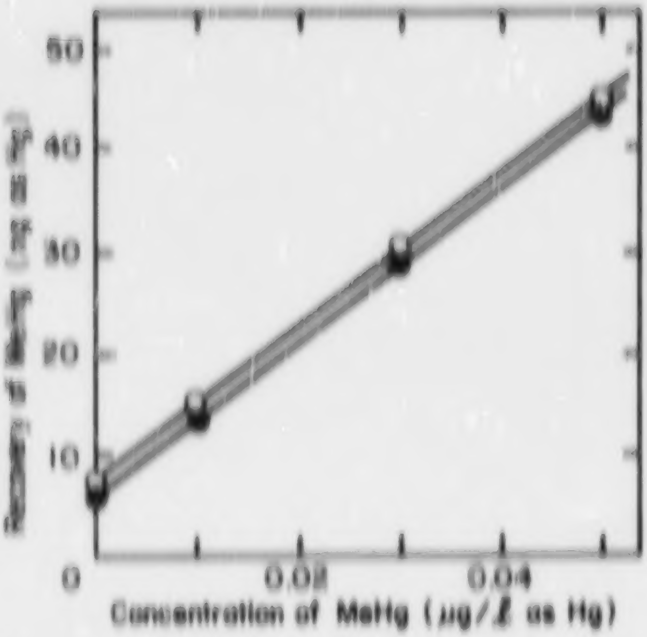


Figure 5. Effect of inorganic mercury on the calibration curve of methylmercury.
◐MeHg and Inorg Hg (0.5 µg/l as Hg) added to deionized water
●MeHg added to deionized water

The calibration curve almost agreed with that of the MeHg solution which did not contain Inorg Hg. It is concluded that Inorg Hg scarcely affected the determination of MeHg in this procedure. Although MeHg and Inorg Hg coexist in samples, and both are adsorbed in chelating resin (A), MeHg is effectively separated from Inorg Hg by subsequent elution and extraction procedure. This is because Inorg Hg is hardly eluted by 4N-HCl, and even if Inorg Hg is eluted slightly, it is scarcely extracted with benzene from 1N-HCl.

Calibration Curve for Determination of Methylmercury in Sea Water

The effect of various ions contained in sea water on the calibration curve of MeHg was investigated. The experimental procedure was the same as the method described previously. The calibration curve for MeHg added to deionized water in the concentration range of 0.01 µg/l to 0.05 µg/l was made at the same time. The calibration curves of MeHg added to the synthetic sea water and the deionized water are shown together in Figure 6.

The calibration curve of MeHg added to the synthetic sea water also had excellent linearity and was in agreement with that of MeHg added to deionized water. In comparing these two calibration curves it is concluded that MeHg can be adsorbed on the chelating resin in the presence of various ions contained in sea water and can then be eluted with 4N-HCl.

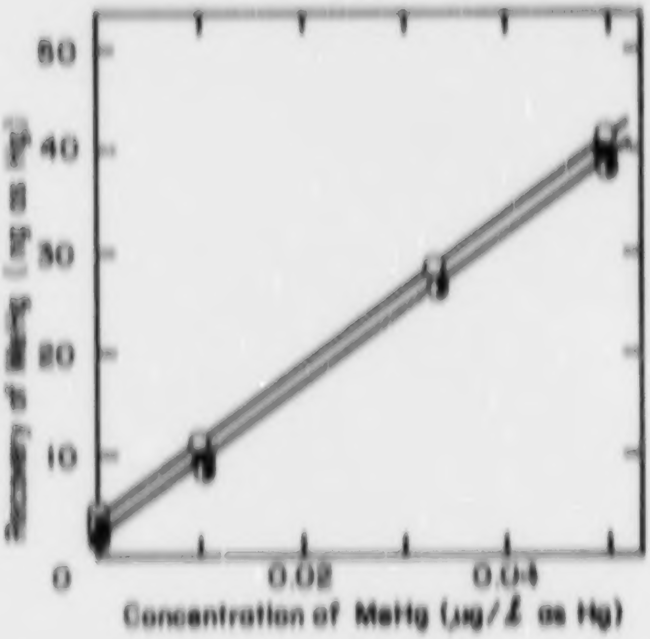


Figure 6. Calibration curve of methylmercury added to sea water* -(I)
●MeHg added to synthetic sea water
○MeHg added to deionized water
*Synthetic sea water prepared by Lyman-Fleming method

The calibration curve of MeHg added to natural sea water in the concentration range of 0.005 μg/l to 0.05 μg/l is shown in Figure 7. The natural sea water was collected from an area which was not contaminated by mercury and was filtered with No. 1 filter paper.

The calibration curve had good agreement when repeated three times. On the basis of the above results it has been found that trace amounts of MeHg contained in sea water can be determined accurately in the concentration range from 0.005 μg/l to 0.05 μg/l.

Determination of Methylmercury in Sea Water Samples

Sampling stations are shown in Figure 8. Sea water samples were collected using a Van Dorn water sampler and filtered with No. 2 paper.

Chelating resin amounting to 0.2 g (A) was added to 1 liter samples of filtered sea water immediately after sampling and all the previous procedures were carried out. The concentration of MeHg in a sea water sample was determined from the calibration curve described previously. The results are presented in Table 2.

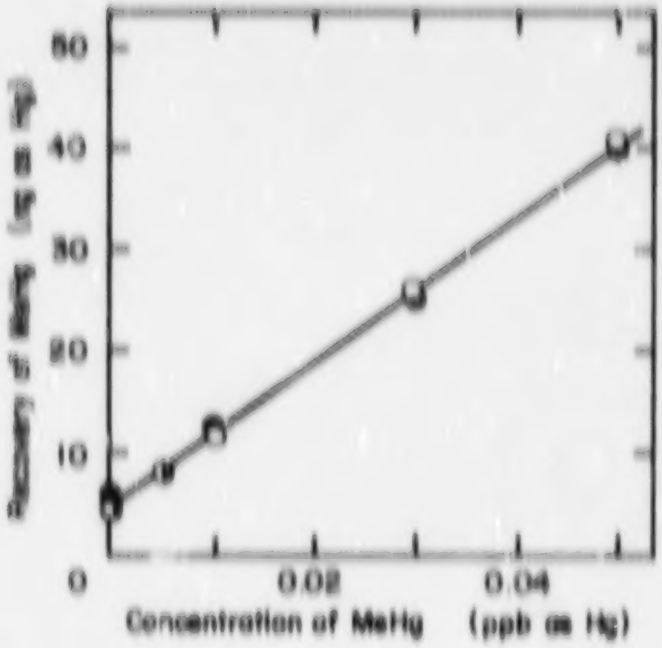


Figure 7. Calibration curve of methylmercury added to sea water* -(II)
● First time ◓ Second time ◒ Third time
*Natural sea water, collected from an area which was not contaminated by mercury

TABLE 2. METHYLMERCURY IN SEA WATER SAMPLES

(Hg μg/l)

Sampling station	Depth from the surface (m)	Date of sampling April 23, 1976	June 3, 1976	August 10, 1976
a	0.5	0.013	0.032	0.005
b	4.5	----	<0.005	<0.005
c	10.0	----	0.010	<0.005

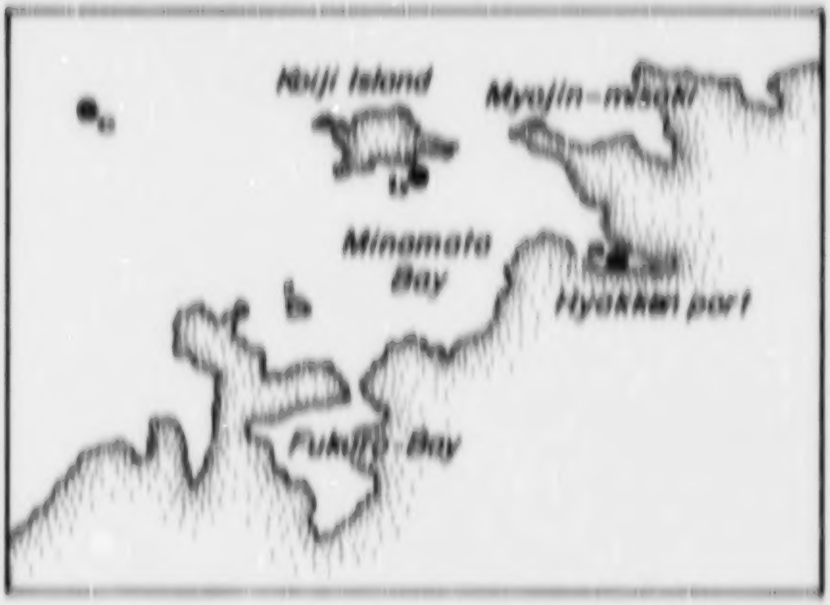

Koiji Island
Myojin-misaki
c
a
Minamata Bay
Hyakken port
b
Fukuro Bay

Figure 6. Sampling stations--a, b, c.

REFERENCES

1) M. Fujiki, R. Hirota and S. Yamaguchi, "Reports of the studies on Minamata Disease," edit. by Japan Public Health Association (1976) p. 16.
2) M. Nishimura, K. Matsunaga and S. Konishi, Bunseki Kagaku, 24, 655 (1975).
3) R. J. Baltisberger and C. L. Knudson, Anal. Chem., 47, 1402 (1975).
4) J. Lyman and R. H. Fleming, J. Marine Research, 3, 134 (1940).

BEHAVIOR OF HEAVY METALS AND PCBS IN DREDGING AND TREATING OF BOTTOM DEPOSITS

K. Murakami* and K. Takeishi**
Public Works Research Institute
Ministry of Construction

ABSTRACT

Many problems concerning water quality arise when bottom deposits containing various toxic substances, such as heavy metals and PCBs, are dredged and treated. These problems are dealt with in this paper, centering around the results of laboratory experiments on the solubility of these toxic substances when contained in bottom deposits and solidified deposits, and the treatment of waste water from processes of handling dredged deposits. The summary of results follows:

(1) The deposit constituents were markedly variable according to particle size, varying with the kind of deposit. The content of heavy metals and PCBs tended to be higher in finer grained sediments.

(2) Mercury and PCBs released from sediments into the overlying water were very low in concentration and most were bound to suspended particles.

(3) Concentration of dissolved PCBs in the wastewater from dredging operations was very low, and dissolved mercury concentration was also low in the normal range of pH. Therefore, it seems that the quality of effluent from dredging operations can be improved to a level which conforms to the effluent standards by eliminating suspended solids.

(4) Elutriate tests showed that mercury transferred from the deposits into the water tended to increase sharply when the solvent was high in pH value. When mercury-containing deposits were solidified with a solidifying agent of the cement-

---

* Chief, Water Quality Section
** Research Sanitary Engineer, Sewage Works Section
5-41-7 Shimo, Kita-ku, Tokyo 115, Japan

lime group, the solubility of mercury measured by the standard elutriate test was greater for solified deposits than that for the orignal deposits. This was because of a rise in pH value of the solvent. However, dissolved PCB obtained by shaking from solidified deposits was very low in concentration, not more than 1 ug/l.

(5) Even if the solvent is high in pH value, the amount of mercury transferred from the deposits to the water could be remarkably lowered by decreasing the water content of the deposits and by oxidizing them.

(6) Redissolved heavy metals from solidified deposits (other than Hg and PCB) were very low in concentration.

INTRODUCTION

Deposition of sediments containing a large amount of toxic substances, such as heavy metals and PCB, has caused a kind of social problem in Japan, and bottom deposit dredging is now being carried out in various places as one measure of water pollution control. One of the most important things in a dredging operation is to prevent the dispersion of toxic substances into the environment in the course of dredging and treating deposits. If the dredging operation is performed without sufficient understanding of the behavior of these toxic substances and without avoiding the dispersion of them into the environment, the possibility exists that the efforts to control pollution would instead cause secondary environmental pollution.

The method of dredging and treating of bottom deposits may differ depending on the characteristics of the deposits to be handled and the geographic and social conditions of the area where the deposition took place. However, a general flow diagram for a dredging operation may be expressed as shown in Figure 1. Dispersion of toxic substances in bottom deposits into the environment, either in dissolved or particulate form, can occur from every handling process shown in the figure.

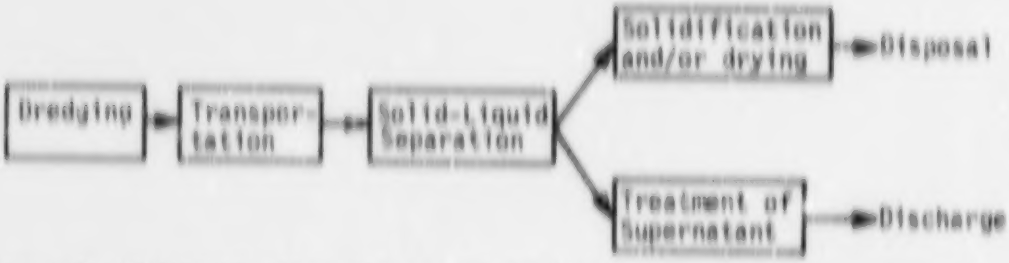


Figure 1. Schematic flow diagram of dredging operation.

These problems are disscussed in this paper with particular emphasis on the dissolving of toxic substances from bottom deposits and the laboratory experiments which are the basis of the results.

CHARACTERISTICS OF THE BOTTOM DEPOSITS USED IN THE EXPERIMENTS

The characteristics of the 4 kinds of deposits used in the experiments are given in Table 1. Sample A (collected from the port of Tagonoura) was from a bottom deposit formed mainly by paper mill waste discharges. Sample B (Minamata Bay) was from a sandy deposit and had relation to chemical industry wastewater discharge. Sample C (Naka no Umi - a salt lake) was related to an electrical industry and fairly low in moisture content. Deposit D (The Tsurumi River) was collected from the tidal reach of an urban river. (Hereafter, samples of these bottom deposits are simply referred to as A, B, C and D).

TABLE 1. CHARACTERISTICS OF THE SEDIMENTS USED IN THE EXPERIMENTS

	Water Content (%)	Ignition Loss (%)	COD (mg/g)	T-P (mg/g)	Kjel-N (mg/g)	PCB or Hg (mg/g)
A	85	44	209.6	2.01	6.57	PCB 111.5
B	70	22	46.06	2.36	1.64	Hg 297.5
C	54	7	20.36	0.29	2.09	PCB 1.0
D	55	16	29.30	3.21	4.12	Hg 0.57

PCB content of Sample A and total mercury content of Sample B were very high and these deposits are the objects of removal and treatment programs according to the "Tentative standards for the removal of deposits containing mercury and PCB" (total mercury: 25 mg/kg; PCB: 10 mg/kg) established by the Environment Agency.

Table 2 shows the concentrations of heavy metals in the deposits and their Clarke number. Sample A contained only 1% Fe in contrast to about 5% as is usually observed in natural conditions, mostly because the components were paper mill wastes. But it contained a considerably higher percentage of Cd and Pb, etc. because of the influence of industrial wastes. Sample B was high in heavy metals in general and had an especially high mercury content, 3,700 times as great as the Clarke number. Sample C was low in content of heavy metals and the value obtained was about the same as the Clarke number, or lower, except for Cd and Hg. The total concentration of 4 heavy metals, Pb, Cr, Cd and Hg, in samples B, A, D and C was respectively, 10.5 times (1213 mg/kg), 6 times, 5 times and 0.6 times as high as that obtained from the Clarke number, indicating that pollution due to these heavy metals was in a remarkably advanced stage in bottom deposit B.

TABLE 2. HEAVY METALS IN THE SEDIMENTS USED IN THE EXPERIMENTS (Hg in mg/kg and others in mg/g)

	Fe	Zn	Mn	Pb	Cu	Cr	Ni	Cd	Hg
A	9.91	1.320	0.106	0.406	0.322	0.128	0.112	0.140	0.527
B	44.10	1.218	0.960	0.537	0.913	0.367	0.140	0.021	298
C	19.93	0.103	0.346	0.029	0.031	0.036	0.047	0.0073	0.440
D	63.0	0.588	0.506	0.438	0.227	0.096	0.094	0.0016	0.567
Clarke Number	48.3	0.060	0.950	0.015	0.055	0.100	0.075	0.002	0.08

CONTENTS OF HEAVY METALS AND PCB BY PARTICLE SIZE GROUP

Each of the bottom deposit samples was divided into classifications by sieving into several groups by particle size, heavy metal content and PCB content to clarify the relation between particle size and Hg and PCB contents. The results are shown in Figures 2 and 3. Table 3 lists the contents of PCB and Hg shown in Figures 2 and 3, and also gives particle size groups as percentage to the total content.

Results for PCB: The total PCB content of Sample A was extraordinarily high. The percentage of PCB content in the particle size group of less than 74 μ was overwhelmingly higher than those in the other groups, i.e., the PCB content of this size group accounted for 99.7% of the total content (Table 3).

Results for Hg: Hg content tended to be higher in finer particles in all the deposit samples except A, and about 90% of the Hg was contained in the particle size group of less than 74 μ in B (Table 3). The concentrations of other heavy metals were also generally higher in the finer particles. But Sample A tended to be different from the other samples in the contents of many heavy metals. There was no definite trend in the distribution of heavy metal content in Sample A. This is probably because most of the constituents of the deposit were specific substances, artificially produced and the heavy metals contained in it were introduced under different conditions when compared with those in the other samples.

Relation among heavy metals: Relatively good correlations were observed between Cu and Zn content and between Cu and Pb content, but there were no notable correlations between the others (Figures 4 and 5).

As mentioned above, toxic substances were in many cases contained in specific particle size groups. Though the particle size which exhibits the highest concentration is variable (depending on the sediment characteristics and the toxic substances) it can be said that these toxic substances as a

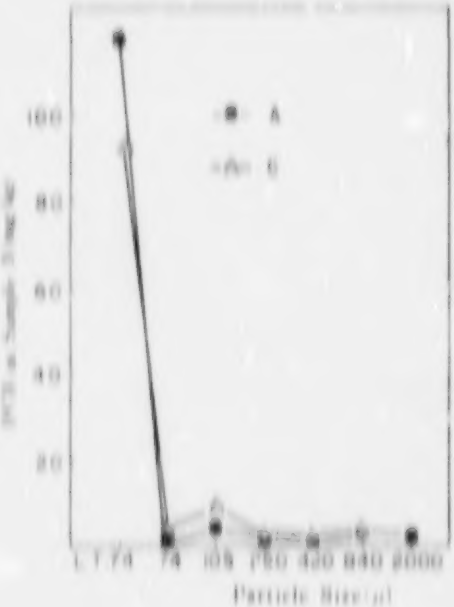


Figure 2. Relationship between PCB content and size of sediment particles.

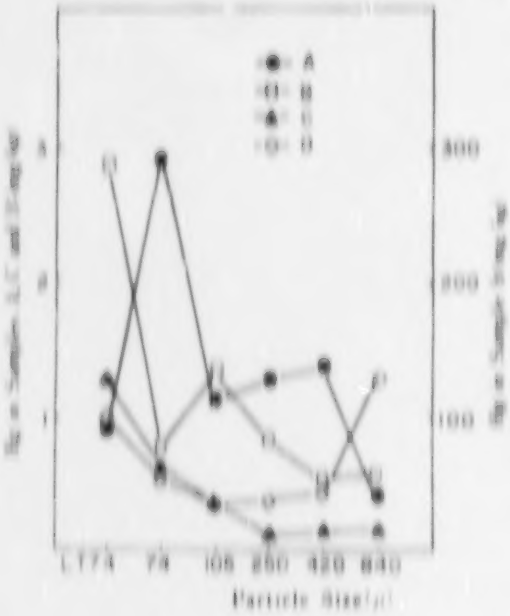


Figure 3. Relationship between Hg content and size of sediment particles.

TABLE 3. SEDIMENT PARTICLE SIZE GROUPS VS. PCB AND HG CONTENT

	Sample A			Sample B		
Particle Size (μ)	Weight Ratio of Solid (%)	PCB Content (mg/kg)	Weight Ratio of PCB (%)	Weight Ratio of Solid (%)	Hg Content (mg/kg)	Weight Ratio of Hg (%)
L.T. 74	86.07	117.0	99.65	75.88	284.5	91.40
74 - 105	1.59	0.69	0.01	0.56	77.2	0.18
105 - 250	3.96	3.99	0.16	4.07	133.0	2.29
250 - 420	1.79	1.41	0.02	12.71	82.9	4.47
420 - 840	1.87	1.73	0.03	2.95	54.0	0.67
840 - 2000	3.34	3.30	0.11	2.76	58.1	0.99
M.T. 2000	0.97	1.98	0.02	0.12	----	----

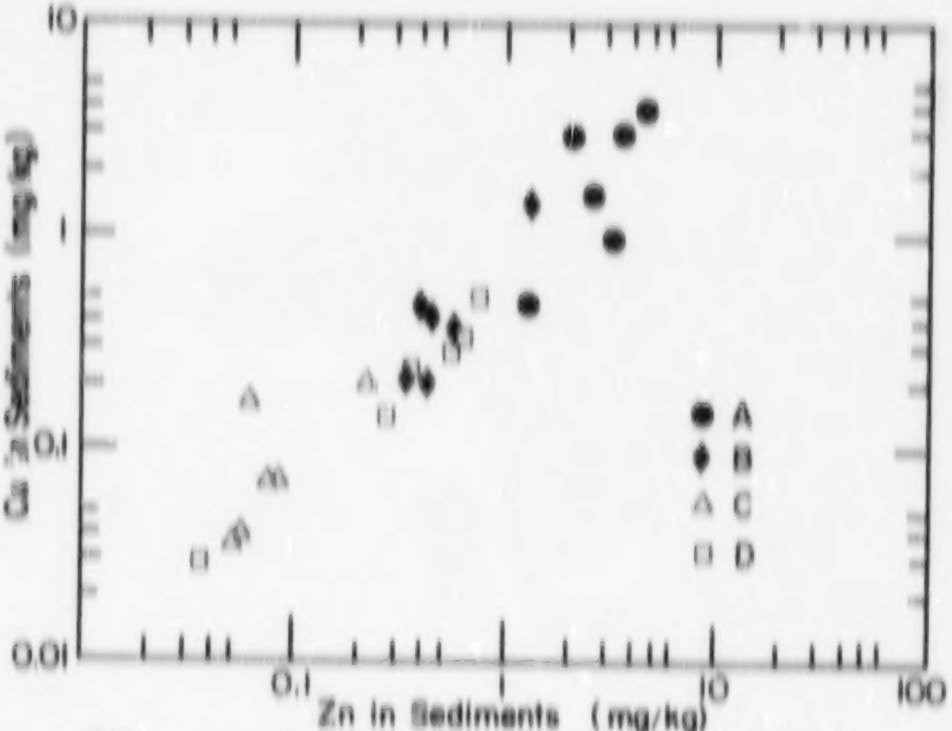


Figure 4. Relationship between Cu and Zn in sediments.

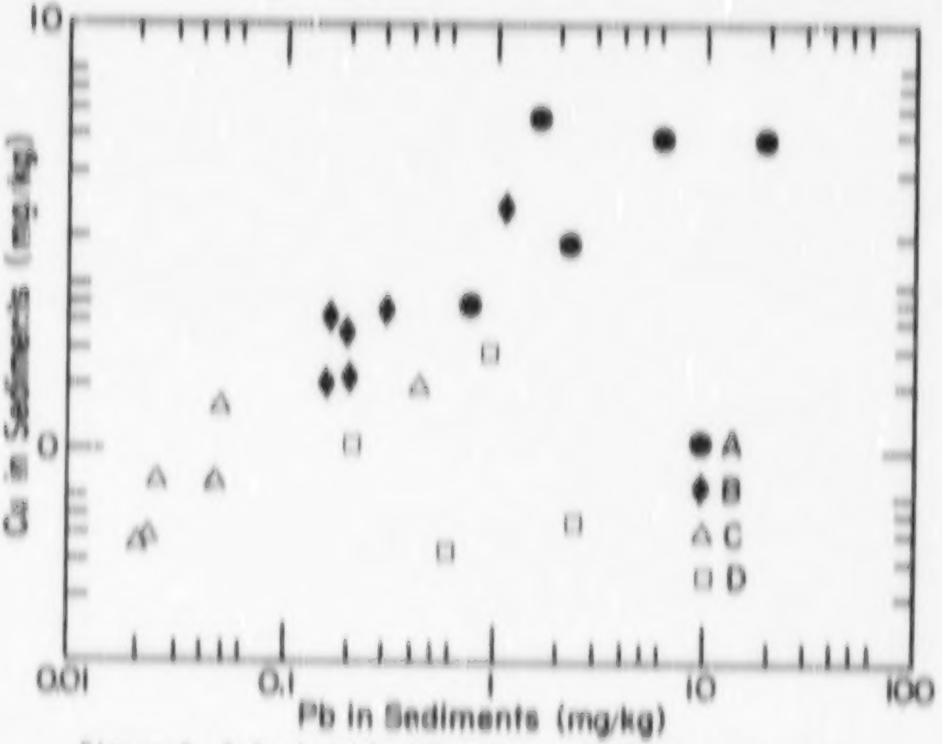


Figure 5. Relationship between Cu and Pb in sediments.

rule have a tendency to be contained in the finer particles of bottom deposits. Therefore, attention must be paid to the fact that resuspension of bottom deposits at the time of dredging and disposal of untreated effluent from handling and treating spoils will cause fine particles of deposits to disperse and will result in environmental pollution due to any toxic substances contained in the dredged deposits.

RELEASE OF HG AND PCBS INTO OVERLYING WATER FROM BOTTOM DEPOSITS

Experimental Method

A 10 cm thick sediment sample was laid at the bottom of each 75 liter cylindrical vessel and either sea water or fresh water was placed over the sediment in the cylinder. The water in each cylinder was kept either under anaerobic conditions by an air-tight seal at the top of the cylinder or kept under aerobic conditions by aerating the water. In both cases the water was slowly mixed to ensure a uniform concentration throughout. All the experiments were carried out at a constant temperature of 20° or 30°C. During the experimental period of 500 days, water samples were regularly taken from each cylinder and analyzed for PCB or Hg as well as pH, ORP, SS, TOC and other parameters.

Sediment samples used in the experiments were types A and C for the experiment on mercury, and types B and D for that on PCB. Other sediment samples collected from rivers were also used after adjusting the PCB or Hg contents.

In addition to the 75 liter-cylinder test, smaller scale experiments were done using 300 ml flasks to supplement the cylinder test. In the flask test a sediment sample was placed in a series of flasks and the flasks were filled with distilled water. These flasks, after being glass-stoppered, were stored in a 20°C constant temperature room and were opened sequentially for water quality analysis to examine the release of PCB or Hg from sediments to overlying water.

Release of PCB

The experiment included twelve 75 liter cylinder tests and ten flask tests. The PCB added to the river sediment samples used in the 75 liter cylinder tests was a mixture of KC (Kaneclor) 300, KC 400, KC 500 and KC 600 with a weight ratio of 1:1:1:1 and the PCB content in the samples was adjusted to 50 or 100 μg/kg (on a dry basis). In the flask test the mixture mentioned above as well as each project (KC 300-600) was used to adjust PCB contents in the samples.

The ranges of pH and ORP variation of the overlying water were respectively 6.5 to 8.5 and -0.4 to 0.1 V. The concentration of PCB in the water decreased successively in most cases with no relation to ORP or pH. In some cases comparatively high concentrations of PCB 30-60 μg/kg) were observed. At these times suspended solids in the water also tended to be high. When the PCB concentrations were corrected (on the assumption that the sediment PCB content in each case was 100 μg/kg) in order to examine the relationship between the PCB and SS in the water, a comparatively good correlation was seen as shown in Figure 6. The solid line in the figure shows the relationship in which all PCB in the water is present in the form of suspended solids. From this figure it was concluded that the resolubilization of PCB from bottom sediments to overlying water is very small.

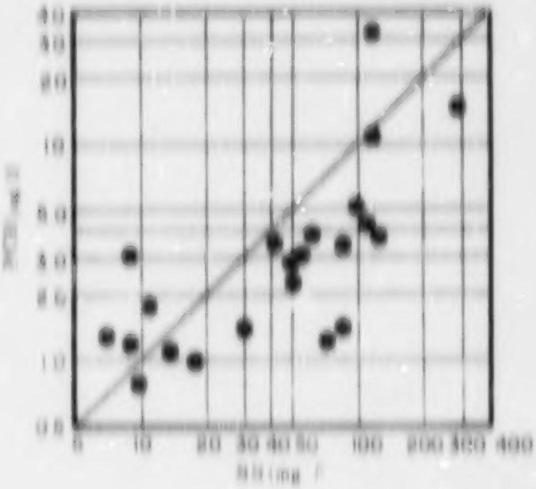


Figure 6. Relationship between PCB and suspended solids in overlying water. PCB concentration in the overlying water is corrected according to the PCB content of bottom deposits: 100 μg PCB/g basis.

Release of Mercury

The experiment included ten 75 liter tests and fifteen flask tests. The form of mercury experimentally added to river sediment samples was either $HgCl_2$ or Hg_2S and the mercury content in sediment samples was adjusted to several concentration levels in the range of 5 to 100 μg/kg. The ranges of pH and ORP were the same as those of the experiments on PCB. In the early stage of the experiment, the concentration of mercury in the overlying water was in some cases as high as 20-30 μg/l because of resuspension of bottom sediments, but the value decreased gradually, becoming less than 0.5 μg/l (detectable limit) in all cases even after about 500 days. It seemed that the concentration of mercury in the overlying water had no specific relation to other water quality parameters such as pH and ORP within the range of these experiments, and the mercury diffused from the deposits in soluble form in a very small amount.

TRANSFER OF HG AND PCB INTO THE SUPERNATANT IN THE DEWATERING OPERATION OF DREDGED DEPOSITS

In order to reduce the contents of Hg and PCB in the supernatant from mechanical dewatering processes, experiments on the treatment of supernatant obtained by the leaf test of Deposits A and B were carried out by chemical coagulation with metal salts and/or a polymer.

The PCB contained in the supernatant of sample A was considered to be mostly bound to suspended solids, as shown later (Figure 9). Therefore, it seemed possible to reduce the concentration of PCB in the supernatant to the level of the effluent standard by the removal of suspended solids.

In the case of Hg, when lime was used as a conditioning chemical for filtration, the soluble Hg concentration tended to increase in the supernatant because of a rise in pH, and the concentration of residual Hg was 10 μg/l or more, even after chemical coagulation. Therefore, the effect of pH on the solubilization of Hg from bottom deposits was further examined using elutriate tests. Deposit sample B was added to distilled water at a concentration of 3 g of solids per 100 ml (on a dry basis) and the pH of the mixture was adjusted to several levels with NaOH or H_2SO_4. After violent shaking the supernatant of each mixture was filtered through a 0.45 μ membrane filter and the filtrate was analyzed to examine the relation between the Hg concentration or COD and the pH value. The results are shown in Figure 7. The pH value of the mixture without pH control was 8.4.

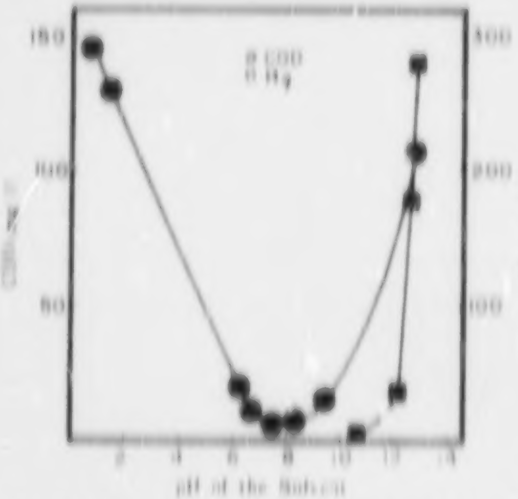


Figure 7. Relationship between solubilization of Hg and COD from the sediments and pH of the solvent. Obtained by the elutriate tests.

The concentration of soluble Hg in the supernatant was sharply increased at about pH 11 and the elutriate ratio reached 3.1% at pH 12.8. Obviously pH value has a great influence on the behavior of mercury contained in deposits. Therefore, changes of pH value should be avoided as far as possible in the treating of dredged spoils.

TREATMENT OF WASTEWATER FROM DEWATERING PROCESSES OF DREDGED DEPOSITS

The deposit dredging operation system now widely in use is as follows: pump dredging ⟶ pipe transportation ⟶ landfill. In this system, the water content of dredged deposits is usually kept at more than 90% because of pump and pipe transportation requirements. Therefore, a large quantity of excess water returns from landfill disposal sites, and when deposits contain heavy metals and PCB, it is especially important to keep the quality of effluent higher than the effluent standard in order to carry out successful spoils disposal. In the present study, the possibility of treating the supernatant containing Hg and PCB was investigated using deposit samples A and B.

Treatment of Wastewater Containing PCB

Sample A was added to distilled water at a concentration of 3 g dry solid/100 ml water and shaken. After a quiescent period, the supernatant was taken and used as the water sample for the experiment on chemical coagulation. PAC (polyaluminum chloride) and/or an anionic polymer were used as coagulants. Figure 8 indicates the relation between the coagulant dosage and the turbidity of the supernatant. The effect of PAC in reducing turbidity was greatest at a dosage of about 1000 mg/l. When the polymer was added to the PAC, the turbidity was at a minimum with a polymer dosage of 10 mg/l.

Figure 9 shows a correlation between the turbidity and the PCB concentration. In this figure, data obtained by chemical clarification of the supernatant from vacuum filtration of deposit sample B are included. The correlation between the turbidity and the PCB content was fairly good. From the fact that a correlation between turbidity and PCB in treated water was observed not only in the samples prepared, by a kind of elutriate test as in this experiment, but also in the interstitial water of deposits, it seems that the influence of the solid content or salinity on the solubilization of PCB is rather slight. The sample water, prepared by a kind of elutriate test as mentioned above, was filtered through several kinds of membrane filters with different pore sizes, and the relation between turbidity and PCB of the filtrate was examined. These results are also shown in Figure 9. When these data are compared with the other data obtained by chemical coagulation, it can be seen that the PCB content tended to be a little higher in the former, suggesting that a part of the soluble PCB was removed by chemical coagulation. If PCB contained in water filtered through a 0.45 μ filter is defined as soluble PCB, then the content of soluble PCB in the supernatant was confirmed by physical stirring to be not more than 3 μg/l under such conditions of solubilization. Therefore, it is considered possible to obtain effluent that conforms to the effluent standard if suspended solids are completely removed by chemical coagulation and/or filtration.

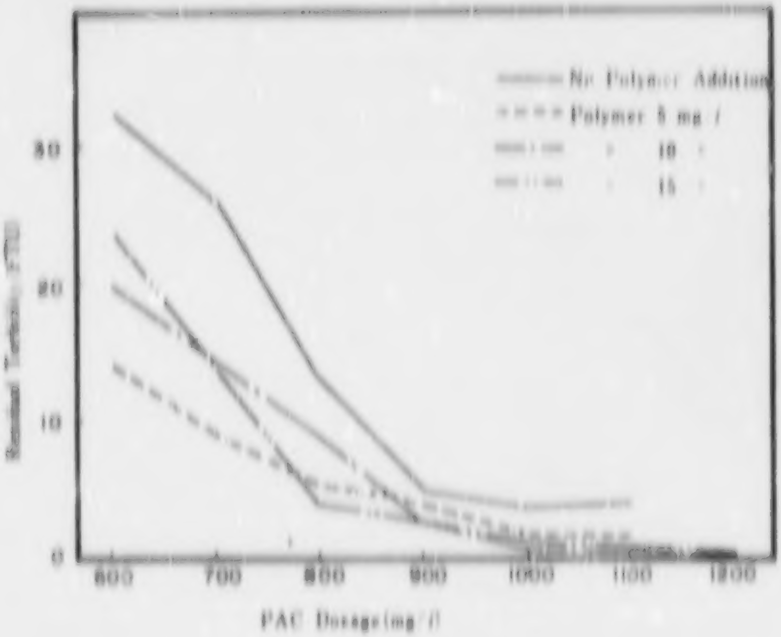


Figure 8. Relationship between residual turbidity and coagulant dosage

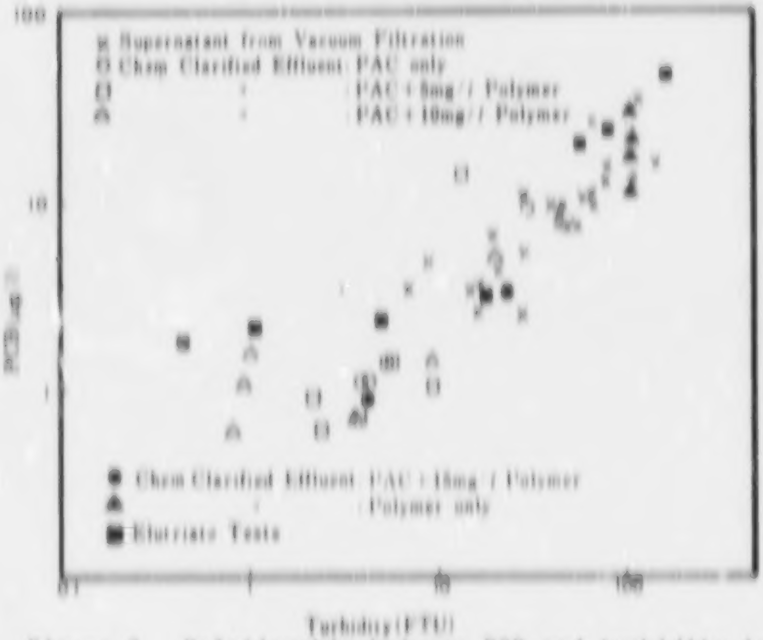


Figure 9. Relationship between PCB and turbidity in various kinds of effluent from bottom deposits.

PCB and its Solubility

PCB used in Japan in the past was almost exclusively Kaneclor, and Kaneclor includes 4 main products, KC 300, KC 400, KC 500 and KC 600. The figures following KC stand for chlorine content; for instance, KC 600 contains 60% chlorine. PCB becomes higher in viscosity and lower in solubility with an increase in chlorine content.

Table 4 shows the solubility of each PCB product, KC 300-KC 600. In Experiment I PCB was dissolved in distilled water by shaking violently, and allowed to stand for 10 minutes. Then samples were taken from the middle part of the water column and used for analysis. In Experiment II PCB was dissolved in the same manner as in Experiment I and, following filtration through a 0.45 μ filter, the filtrate was used for analysis.

TABLE 4. SOLUBILIZATION OF PCB

	Experiment I	Experiment II
KC-300	0.092 mg/l	0.068 mg/l
KC-400	0.072	
KC-500	0.018	
KC-600	0.012	0.006

As mentioned above, the solubilization of PCB from deposits is very small and most of the PCB transferred to the water phase is considered bound to suspended solids. But the solubility of PCB itself as shown in Table 4 is fairly high, much higher than the level of the effluent standard. Therefore, depending on the characteristics of the deposits, there may be a possibility that more PCB is resolubilized.

PCB is a mixture of various chlorinated biphenyls and exhibits 29 peaks on a gas-chromatogram of 9 chlorinated compounds with 2-9 chlorines. Figure 10 indicates the percentages of various chlorinated compounds in two products, KC 300 and KC 600, and in their water soluable fractions. The composition of the water soluble fraction are those with lower chlorine numbers and the composition of the water soluble fraction is considerably different from that of KC 600 itself. That means the solubility of PCB is higher in compounds with lower chlorine numbers than in those with higher chlorine numbers.

Treatment of Mercury-Containing Wastewater

Deposit sample B contained much sand and the quality of the supernatant obtained by elution of the deposits was fairly clean. Accordingly, when treated by chemical coagulation in the same manner as in the case of sample A, the Hg concentration of the supernatant could easily be decreased by the addition of an inorganic coagulant (several tens of μg per liter) to a level

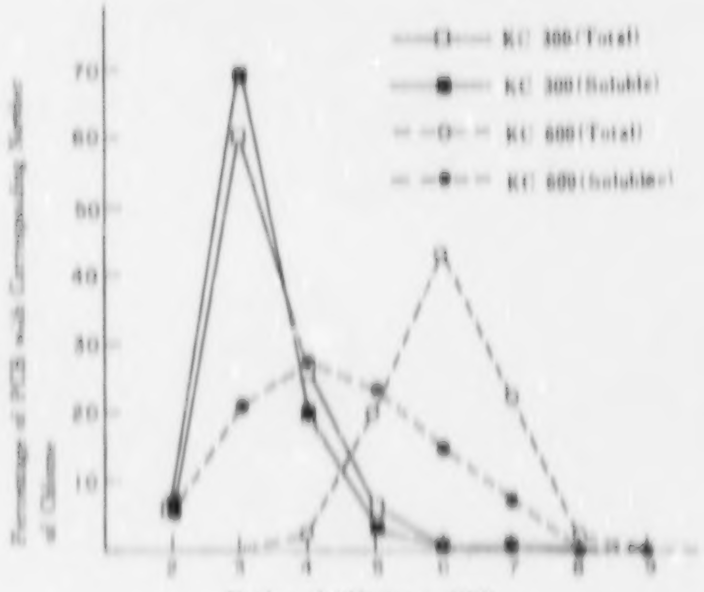


Figure 10. Composition of PCBs with respect to number of chlorine.

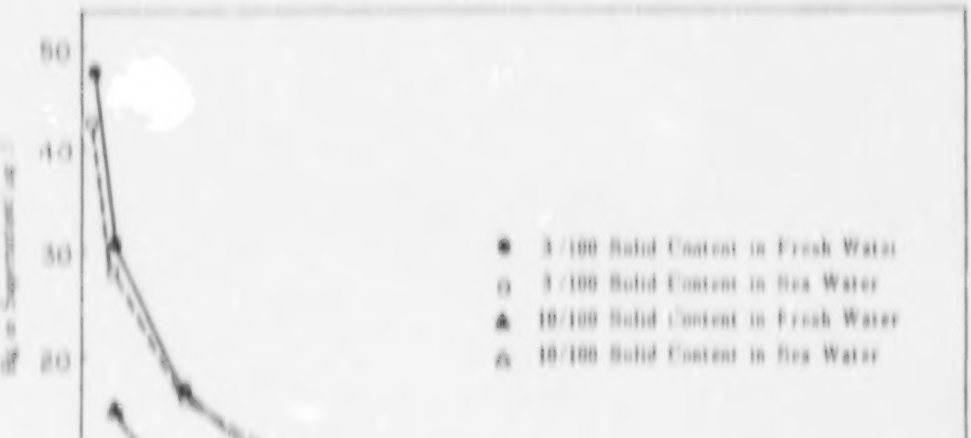


Figure 11. Relationship between Hg in the supernatant and period of settling (ordinary sedimentation).

lower than the detection limit (0.5 μg/l). Therefore, treatment by ordinary sedimentation was also examined. Deposits were added to sea water or distilled water at a solid content of 10 g/100 ml and 3 g/100 ml (dry deposit/water) and, after shaking, changes of the quality of the supernatant with settling time were examined to examine the efficiency of the gravity treatment consolidation alone.

The relationship between the settling time and the Hg concentrations of the supernatant is shown in Figure 11. The quality of the supernatant seemed to be poorer with sea water than with distilled water, probably because of the lower settleability of particles in the former. But in all the cases the Hg concentration could be decreased to a lower level than the effluent standard by 24 hours quiescent settling.

Figure 12 shows the relationship between SS and Hg in the supernatant as observed in the above settling test. This figure indicates that there is little influence by salinity or solid content on the concentration of soluble Hg in the supernatant. There was a fairly good correlation between the SS and the Hg content. The solid line in the figure indicates that all the mercury in the water is included in suspended solids, which are the solids of deposit sample B. Data in the figure agree fairly well with the solid line. Therefore, it is assumed that almost all the Hg exists bound to suspended solids and its solubility in water is very low under conditions of physical stirring.

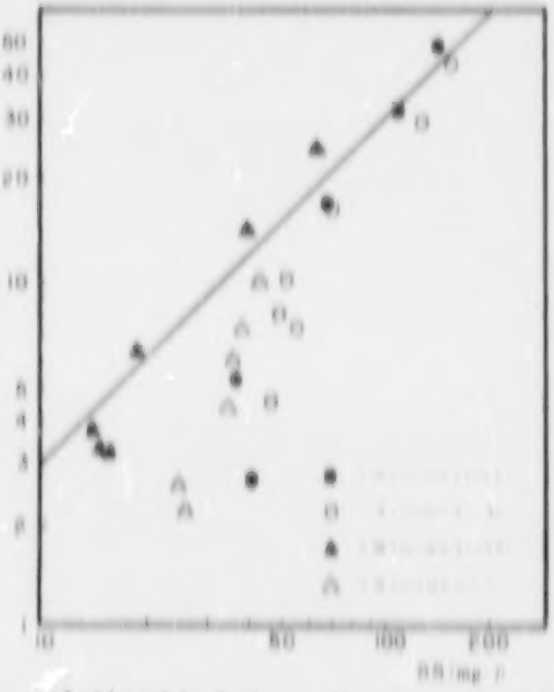


Figure 12. Relationship between Hg and turbidity in supernatants obtained by elutriate tests.

SOLUBILIZATION OF TOXIC SUBSTANCES FROM SOLIDIFIED DEPOSITS

Disposal of deposits containing toxic substances such as heavy metals is provided for in the "Ordinance of the Prime Minister's Office on the standards for judgment of harmful industrial solid wastes--Tests for toxic substances contained in industrial solid waste" (February, 1973). Elutriate tests for toxic substances should be performed before landfill disposal. If the elutriate ratio is higher than the standard, then measures must be taken to prevent toxic substances from diffusing out when deposits are solidified with cement, etc. In this section, the results of experiments on solubilization of toxic substances from solidified deposits are discussed. Sediment samples A and B were used for this experiment.

Solubilization of Toxic Substances from Solidified and Crushed Deposits

Sediment samples were mixed with several separate kinds of solidifying agents, and each of them was put into a 5 × 10 cm form and cured in wet air at 20°C for 7 days. The solidified deposit obtained was subjected to an unconfined compression strength test and then crushed into particles according to the "Tests for toxic substances contained in industrial solid wastes." The particles were sieved, and the ones with a size of 0.5-5 mm were placed in distilled water adjusted to pH 7.8-8.3 with NaOH. The solid content was adjusted to 10 g dry solids/100 ml water). The mixture was shaken for 6 hours and the supernatant was filtered through 5C filter paper; the filtrate was used as the sample for analysis.

Some of the results obtained by the above mentioned standard elutriate test are given in Table 5. These data were obtained from solidified deposits of about 1 kg/cm^2 in strength. The elutriate ratio is defined as the ratio of the amount of a substance dissolved in the water to the amount present in the deposit.

The pH of the solvent (distilled water) increased with the amount of solidifying agent added. The solubilization of PCB from solidified deposits was very low--concentration in the filtrate was between 0.3 and 1.0 μg/l. The concentrations of Cd, Cr and Pb in the filtrate were also low--at or below the detection limit in almost all cases--and showed that an increase in pH did not affect the solubility of these metals. However, the concentration of Hg was considerabluy higher (0.7-144 μg/l) when compared with the other heavy metals and with the standard for Hg concentration (5 μg/l). This increased Hg concentration is probably due to the increased pH of the solvent.

Figure 13 shows changes with curing time of the soluble Hg concentration in the filtrate. The data on the zero day of curing were obtained by performing the elutriate test immediately after mixing in the cement. The solubilization of Hg showed a strong decline after 3 days curing, but declined more slowly after that, until the change became practically nil after about 30 days of curing. The cement dosage affected the solubilization of mercury by increasing solubilization with an increase in the cement dosage while the dosage was low. However, as the dosage became high, the solubilization reached the maximum rate, and then decreased gradually with the increase in the dosage.

TABLE 5. RESULT OF ELUTRIATE TESTS USING SOLIDIFIED AND CRUSHED DEPOSITS
(Figures in parentheses are elution ratios in percentages)

Name of Deposit Sample	Initial Water Content of Deposit (%)	Solidifying Agent and Dosage (g/100g dry solid)		Compressive Strength of Solidified Deposit (kg/cm)	pH	COD (mg/l)	Cd (mg/l)	Pb (mg/l)	Cr (mg/l)	Hg (μg/l)	PCB (μg/l)
A	54.5	Cement	20	0.97	10.0	114 (1.30)					0.9 (0.002)
A	64.3	Cement	40	1.00	11.7	155 (2.37)					1.8 (0.03)
A	65.5	Slaked Lime Sand	30 190	0.89	12.7	180 (2.46)					0.5 (0.02)
A	65.5	Quick Lime Sand	40 130	1.03	12.7	101 (2.07)					Tr. (-)
B	56.6	Cement	20	5.98	12.1	203 (11.0)	N.D. (-)	N.D. (-)	N.D. (-)	22.4 (0.19)	
B	30.5	Cement	20	1.17	11.4	63 (2.5)	0.001 (0.24)	N.D. (-)	0.02 (0.19)	N.D. (-)	
B	56.6	Slaked Lime	80	1.73	13.0	179 (12.0)	N.D. (-)	N.D. (-)	N.D. (-)	35.6 (0.37)	
B	56.6	Quick Lime	45	0.65	1.29	214 (12.6)	N.D. (-)	N.D. (-)	N.D. (-)	83.4 (0.75)	

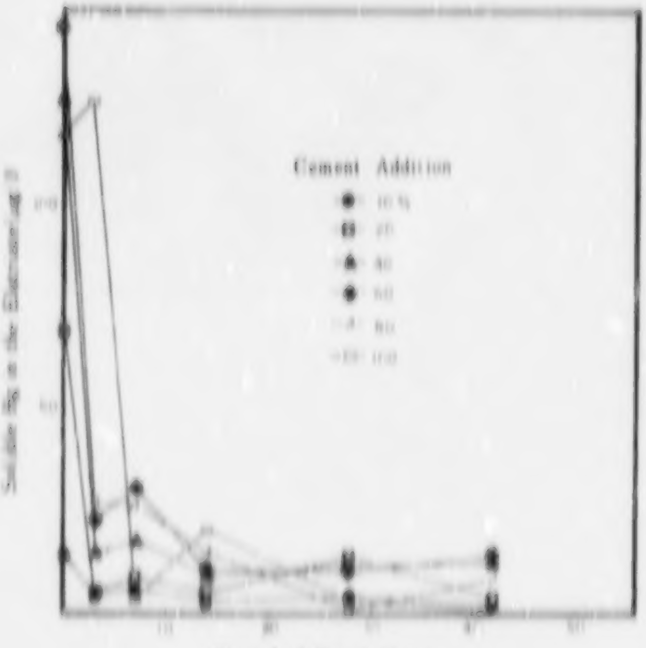


Figure 13. Effect of curing period on resolubilization of mercury from solidified sediments (Standard Elutriate Test).

Although deposit sample B has a very high Hg content of 300 mg/kg, when an elutriate test was performed without solidification the soluble Hg in the filtrate was only 2 µg/l. Therefore, solidification by cement or lime is meaningless as far as the solubility of Hg per unit surface area is concerned. The effect of solidifying agents which raise the pH value of a solvent should only be regarded as a physical fixing of the deposits themselves.

Solubility of Toxic Substances from Bulk Solidified Sediments

The solubilization of toxic substances from bulk solidified sediments was investigated by two kinds of experiments:

Experiment I: The sediment (Sample A) was mixed with cement to form solidified spherules weighing 67-07 g. Each solidified spherule contained 25 g of deposit of a dry basis. Several solidified spherules prepared under the same conditions were soaked in distilled water in a series of 2 liter glass bottles. Water in each bottle was analyzed sequentially after predetermined periods, and the solubility of toxic substances was investigated.

Experiment II: Cement-solidified deposit cubes weighing 320-380 g were prepared using samples A and B. Each cube was soaked in 40 liters of distilled water or sea water for about 40 days. The solubility of toxic substances was examined by taking water samples during the experimental period.

In the tests with sample A (Experiment I) the pH of the solvent (water) increased from to 10 to 12 in the early stages and then decreased gradually due to the absorption of CO_2 from the atmosphere. The increase in pH seemed to be influenced by the constituents of each solidified cube i.e., if the cement content was fixed, the increase of pH was greater when the water content was higher and the strength of the cubes was lower. When the water content was fixed, the pH increase was greater with a high cement content in the solidified deposits.

The concentration of PCB in the solvent was less than the limit of detection (0.1-0.6 μg/l), except for one case which showed a maximum value of 1.1 μg/l. As PCB is a very stable compound, and difficult to influence chemically, it is probable that solidification is effective in reducing the solubility of PCB.

In Experiment II the pH value of the solvent gradually increased for about 20 days, but the increase was especially sharp in the first 5 days. The ORP of the solvent tended to be low when the pH was high, but there was not a definite trend.

In considering the solubility of heavy metals, Cd seems to have diffused out a little from solidified Deposit A, but its concentration was less than the detection limit in the other experiments. Therefore, it may safely be said that Cd is hardly solubilized under these conditions. The concentrations of T-Cr and Hg were hardly more than the detection limit; the determination of Mn, Fe, Cu and Ni were performed only on the last day of the experimental period, but none of them could be detected in the solvent with the exception that Cu content was 0.01-0.18 mg/l in Deposit A, and Fe content was about 0.25 mg/l in both A and B when samples were soaked in sea water.

As mentioned earlier, in the elutriate test using solidified crushed deposits the Hg content of solvent tended to increase with a rise of pH value. But, in the soaking test of bulk solidified deposits, the solidification was effective for confinement of Hg, and even if the pH of the solvent was increased, no notable solubilization of Hg was observed.

Factors Affecting the Solubilization of Hg in Deposits

In ordinary practice the bottom deposits are dredged, and then dried and oxidized under the conditions of landfill disposal. In order to investigate the effect of drying on the solubilization of Hg, deposit samples were air-dried and the solubilization of Hg was then measured using elutriate tests. In addition, the effect of solvent pH on the solubilization of Hg was again examined in order to confirm the results mentioned above. The results are shown in Table 6.

The solubilization of Hg evidently increased with a rise in pH. This was especially true when the pH was adjusted with NaOH and the deposits entered a colloidal state as a result of which the solubilization of Hg was remarkably increased. But, if deposits were dried before treatment, the solubilization of Hg was noticeably low, and this phenomenon was especially remarkable at high pH values. Therefore, a fixed amount of $Ca(OH)_2$ was added

TABLE 6. EFFECT OF DRYING OF DEPOSITS ON SOLUBILIZATION OF HEAVY METALS
(Obtained by elutriate tests. Figures in the parentheses are elution ratios in percentages)

Name of Deposit Sample	Water Content of Deposit (%)	pH Condition of Solvent	pH	COD (mg/l)	Cd (mg/l)	Pb (mg/l)	Cr (mg/l)	Hg (μg/l)
A	85.0	pH 8 by NaOH	7.7	52.7 (0.25)	0.005 (0.04)	N.D. (-)	N.D. (-)	N.D. (-)
A	85.0	pH 13 by NaOH	13.1	2710	N.D.	0.04	0.06	3.9
A	85.0	pH 13 by $Ca(OH)_2$	12.1	123	N.D.	N.D.	N.D	0.6
A	21.1	pH 8 by NaOH	5.5	87.4	N.D.	0.02	N.D.	N.D.
A	21.1	pH 13 by NaOH	13.0	1840	0.004	0.04	0.06	5.2
A	21.1	pH 13 by $Ca(OH)_2$	12.8	113	N.D.	0.01	N.D.	N.D.
B	57.0	pH 8 by NaOH	9.0	16.8 (0.37)	N.D. (-)	N.D. (-)	N.D. (-)	2.0 (0.01)
B	57.0	pH 13 by NaOH	11.3	181	0.005	0.654	0.160	129
B	57.0	pH 13 by $Ca(OH)_2$	12.7	109	N.D.	N.D.	N.D.	148
B	6.96	pH 8 by NaOH	8.0	80.4	0.001	Tr.	N.D.	0.8
B	6.96	pH 13 by NaOH	12.7	167	N.D.	0.02	0.01	3.5
B	6.96	pH 13 by $Ca(OH)_2$	12.8	72.5	N.D.	0.13	N.D.	0.7

to each of several deposit samples (which were different in water content from each other) and the solubilization of Hg from each sample was examined by elutriate test. The intent was to observe the relationship between the degree of drying of the deposits and the solubilization of Hg from them. The results are shown in Table 7. The pH of the solvent was not greatly influenced by drying, but the ORP changed markedly from the oxidized state to the reduced state. The solubilization of Hg decreased sharply with the decrease of water content. From this result, and from the fact that Hg did not dissolve out of solidified-crushed deposits with an initial water content of 30.5% (Table 5), the conclusion is that drying of deposits seems to be greatly effective for decreasing the solubilization of Hg.

TABLE 7. EFFECT OF DRYING DEPOSITS ON SOLUBILIZATION OF MERCURY (obtained by elutriate tests)

Initial Water Content of Deposit (%)	pH	ORP (mV)	COD (mg/l)	Hg (μg/l)
6.83	11.8	+144	104	0.9
14.15	11.9	+123	141	0.3
23.93	11.8	+128	110	0.4
36.61	11.7	-233	126	35.2
54.69	12.2	-214	106	44.9

Diffusion of Heavy Metals Added to Deposits

As mentioned above, the solubilization of heavy metals from natural bottom deposits was very generally low and that of Hg was also low unless it was influenced by some specific factor such as an increase in pH. In order to investigate the heavy metal-stabilizing capacity of deposits, soluble heavy metals were added to deposits and then the transfer of the metals into the water phase examined using the elutriate test.

A mixed solution of $Cd(NO_3)_2$, $K_2Cr_2O_7$ and $HgCl_2$ or a solution of $Pb(NO_3)_2$ was added to deposits A and B at a content of 100 mg/kg (mg of each metal/kg dry solid). Samples were taken at 10 day intervals and the transfer of each heavy metal into the water phase was examined using the elutriate test.

Analysis was carried out 4 times at intervals of 10 days, but the soluble metals in the water phase were always less than the detection limits. This result indicates that every soluble heavy metal added was changed to a stable and insoluble form in the deposits.

A STUDY ON THE BEHAVIOR OF MERCURY-CONTAMINATED SEDIMENTS IN MINAMATA BAY

T. Yoshida* and Y. Ikegaki**

ABSTRACT

This study was undertaken by the Bottom Sediments Management Association, on the request of Kumamoto Prefecture, to investigate the behavior of mercury-contaminated sediments in Minamata Bay in regards to the plan to remove them by dredging. This plan for dredging and disposal of contaminated sediments in Minamata Bay was presented by Mr. Sakomi at the meeting last year in Corvallis, Oregon, USA, (Figure 1). This year it was decided to put the project into practice beginning in 1978. But there are still some technical problems to be solved before the removal operations can begin. One of them is how to treat the toxic metal compounds contained in the sediments, especially mercury compounds, without causing any adverse effects on the surrounding environment. Several tests were conducted on the behavior of mercury compounds in sediments during the dredging and disposal operation. This paper describes the test results and methods of non-polluting treatment of mercury compounds in Minamata Bay dredge spoils.

INTRODUCTION

The sediments in Minamata Bay are heavily contaminated by mercury compounds which were deposited from the waste water discharged from the Chisso chemical factory. Mercury chloride or mercury sulfate was used there as a catalyst for the synthesizing of vinyl chloride or acetaldehyde. After the outbreak of the famous Minamata disease, there has been much investigation of the cause of the disease, the formation of organic and inorganic mercury in chemical reactions or in the sediments, and the transition mechanism of methylmercury from fish to the human body. But there have been few studies on the behavior of mercury in sediments from the standpoint of sediment treatment. As the sediments are dredged and disposed, the metallic mercury in them will become subject to new conditions, i.e., dredging, pumping, settling ponds, land fills, etc. How does mercury behave in these new conditions? To find out, the authors conducted a series of sediment treatment tests and observed the behavior of mercury compounds under these conditions.

---

* Japan Bottom Sediments Management Association, Sankyo Bldg. 2-7-12 Tsukiji, Chuo-ku, Tokyo 104, Japan

**Environmental Department, Kumamoto Pref. 6-18-1 Suizenji, Kumamoto 862, Japan

SAMPLING OF SEDIMENTS

The samples used for tests were taken from the sea bottom outside of the planned embankment, as shown in Figure 1. The sampling was done by hand with two divers, using a 50 ton barge, 150 ps tug boat and 30 ps diver boat. The two divers picked up predetermined quantities of sediments taking care that the samples not be diluted, and filled up large drums with them. The drums were covered and sealed at the sea bottom and then pulled up (Figure 2).

The ocean depth at the sampling site was 10-12 m and bottom soil was sampled between the surface and 80 cm in depth from the surface. The color of the soil was blackish to 20 cm depth and grayish below 20 cm.

ANALYSIS OF PHYSICAL AND CHEMICAL CHARACTERISTICS OF SEDIMENTS

The samples were analyzed according to the following authorized methods.

i) Soil test standards by the Japanese Society of Soil Mechanics and Foundation Engineering.
ii) Guideline "Provisional Criteria for the Removal of Bottom Sediments"
iii) Japanese Industrial Standards

The analytical results of the sediment tests are shown in Table 1. As shown, about 95% of the sediment composition consists of silt and clay, and large quantities of toxic materials as well as mercury are contained in it.

MICROSCOPIC OBSERVATION OF THE SEDIMENTS

The sample sediment was diluted to 350 mg/l in SS (suspended solids) concentration and then, by using a concentric settler, it was divided into 4 fractions according to particle sizes.

As shown in Figure 3, the fractions were microscopically observed.

The grain sizes of the fractions are 100 μ (first), 30 μ (second), 13 μ (third) and 4 μ (fourth). The first one is mostly sand particles, and the second for the most part consists of dead organisms or their fragments. The third contains many salients, and the fourth is a structure mixed with white flakes and needle-shaped particles.

The electron micrographs at magnifications of 1,000X and 10,000X are shown in Figure 4. In these the "flowers" of soil mineral are visible.

X-RAY DIFFRACTION ANALYSIS OF SEDIMENTS

The samples were also observed by X-ray diffraction, and a typical result is shown in Figure 5. In the fourth fraction, with a particle size of 4 μ, the presence of Feldspar and Montmorillonite is recognized and also mercuric sulfite, lead sulfite and mercuric sulfate are detectable.

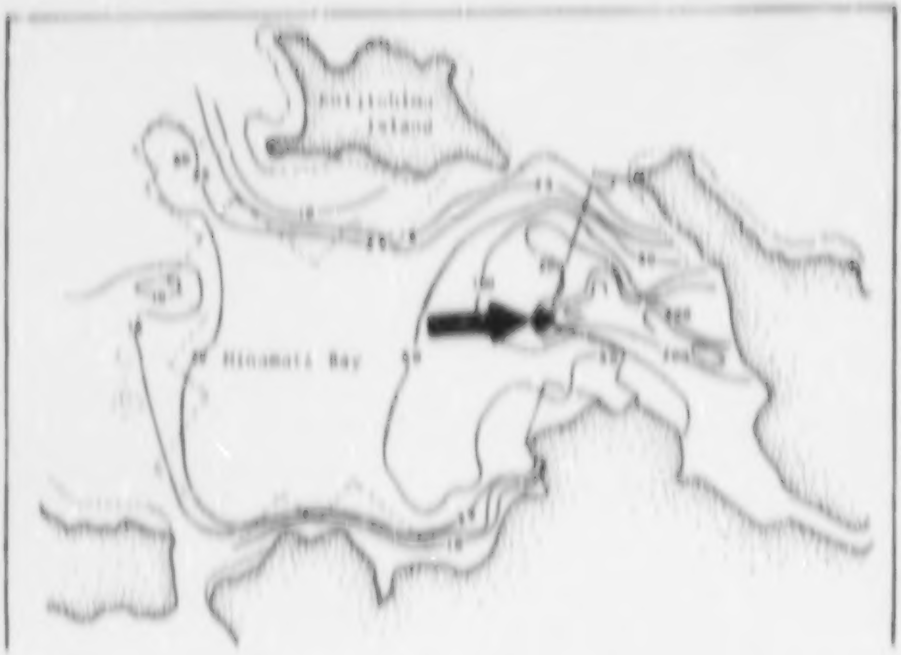
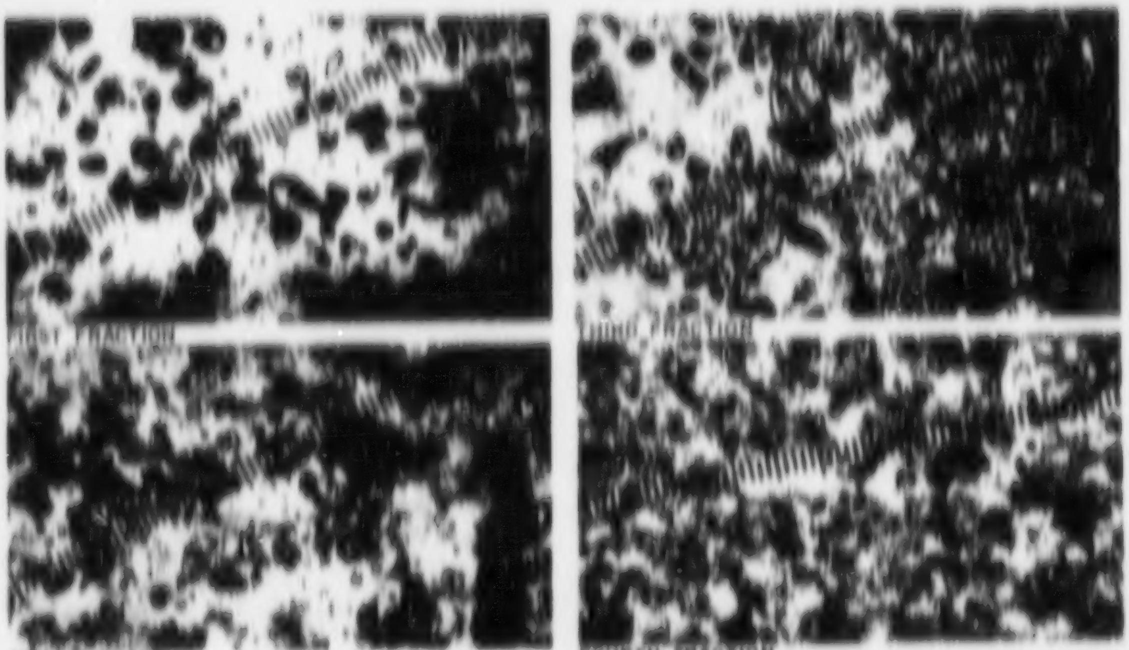

Island
Minamata Bay

Figure 1. Sampling site.

Figure 2. View of sampling.

TABLE 1. PROPERTIES OF SEDIMENT

Item		Unit	Measured Values
Color			Gray blackish
Odor			
pH	(H_2O)		8.13
	(KCl)		8.06
Water Content	$\frac{\text{wt. (g) of water}}{\text{wt. (g) of SS}} \times 100\%$		292
Specific gravity			2.578
Weight of unit volume		g/cm^3	1.16
Ignition loss		%	18.2
Grain size	Max.	µ	420
	D_{60}	µ	22
	D_{30}	µ	12
Total mercury		mg/kg	231
Alkyl mercury		mg/kg	0.003
		"	"
Cadmium		"	1.7
Lead		"	238
Arsenic		"	65
Total nitrogen		"	1750
Total phosphorus		"	928
Organic phosphorus		"	<0.01
PCB		"	0.01
Cyanogen		"	0.3
Sulfide		"	1.5
N-Hexan extracts		"	1.52
Inorganic Constituents	Al_2O_3	%	6.0
	SiO_2	"	36.2
	Na_2O	"	4.3
	CaO	"	9.7
	MgO	"	9.5
	Fe_2O_3	"	2.9
	SO_3	"	5.7
	CO_2	"	2.4
	CuO	"	trace
	P_2O_5	"	"
	ZnO	"	"
	MnO	"	"



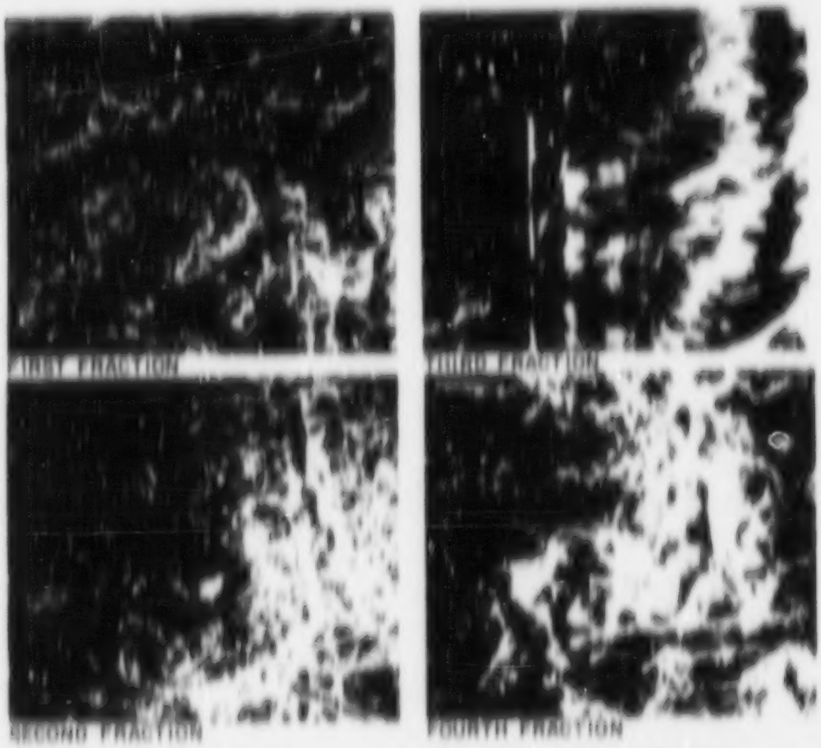
FIRST FRACTION

THIRD FRACTION

SECOND FRACTION

FOURTH FRACTION

Figure 3. Micrographs of Sediments



FIRST FRACTION

THIRD FRACTION

SECOND FRACTION

FOURTH FRACTION

Figure 4. Electron micrographs of sediments.

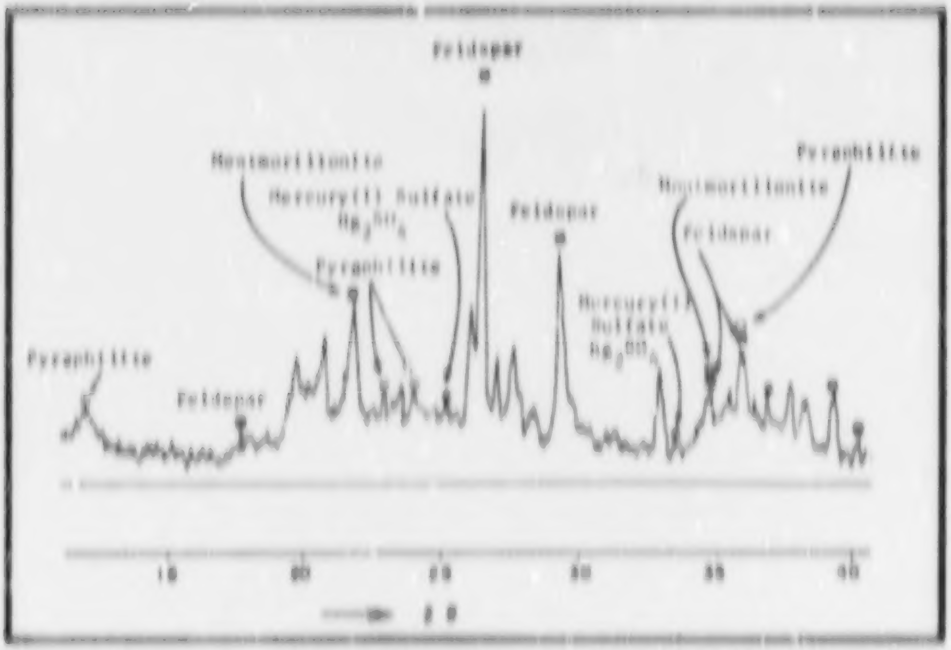


Figure 5. X ray diffraction spectrum of sediment.

BEHAVIOR OF MERCURY BY PHYSICAL ACTIONS

Settling by Gravitation

The sample sediment was diluted with the artificial sea water "Aquamarine" into 8 kinds of SS concentration which were about 50, 100, 500, 1,000, 5,000, 10,000, 50,000 and 100,000 ppm. A 2 liter cylinder was used for the gravity settling of each sample. Figure 6 shows the relationships between settling time and total Hg concentration and also between settling time and turbidity of the supernatant in the cylinder.

The relationship between the residual content of total mercury in the supernatant and the intial concentration of SS was shown in Figure 7. A residual total Hg, defined as the ratio of the total-Hg concentration in the supernatant to that in the sample, was also plotted against the initial SS concentration in Figure 7. From these results it was noted that (1) the larger the degree of dilution, the greater the residual total-Hg and SS, and (2) the maximum concentrations of total-Hg and turbidity in the supernatant were observed in a range of from 1,000 mg/l to 5,000 mg/l of the SS concentration in diluted samples.

Mercury Behavior under Mechanical Agitation of Sediment

A mixture of sediment and Aquamarine, the volumetric percentages of which were 10% and 90% respectively, was prepared and 40 liters were agitated

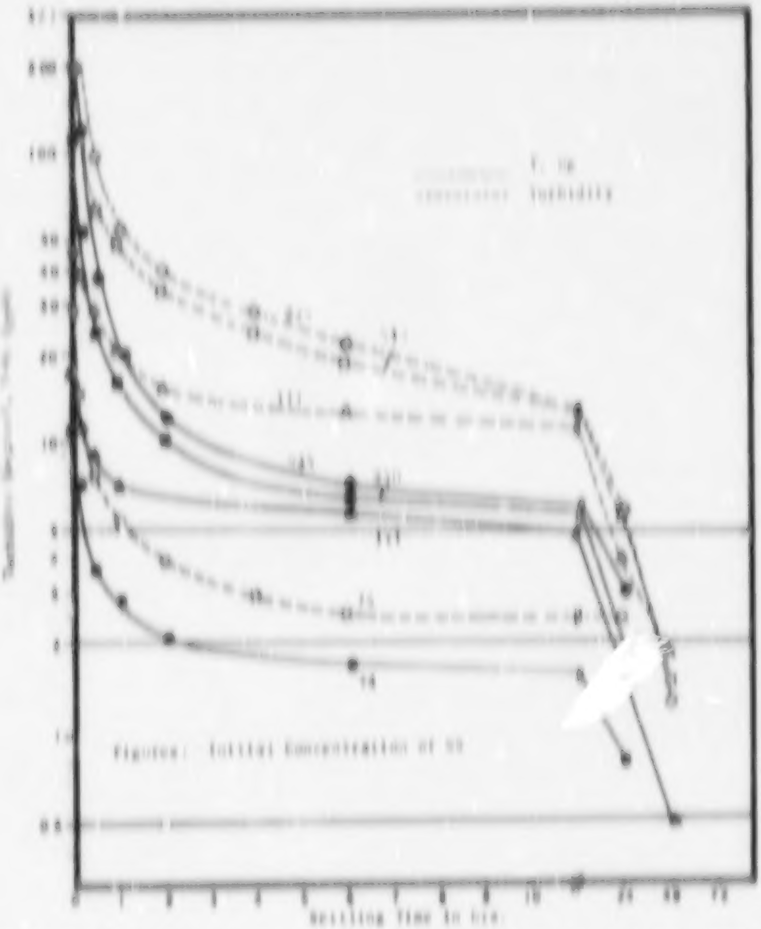


Figure 6. Relationships between settling time and T.Hg, and turbidity.

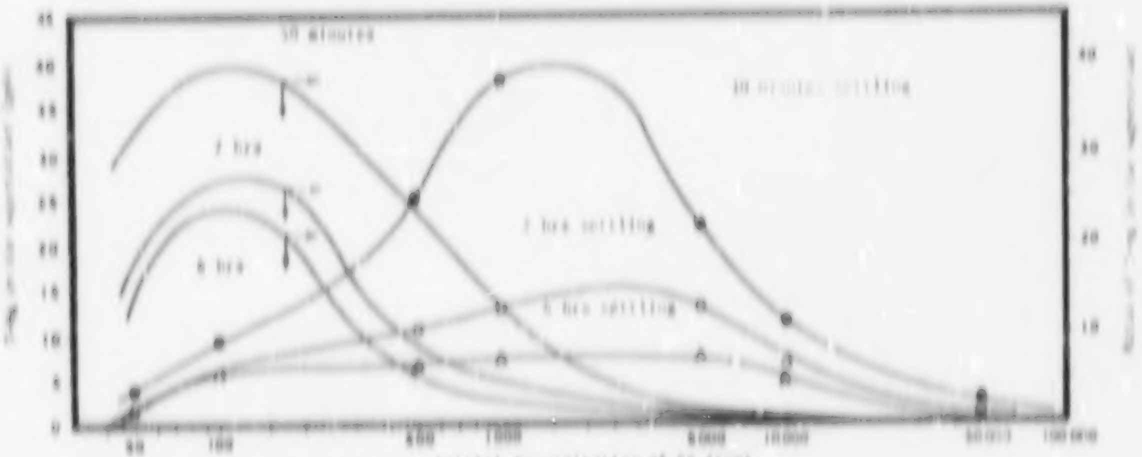


Figure 7. Plots of T.Hg conc. and residual T.Hg against initial conc. of SS.

for 60 minutes by a hand mixer with turbine blades in a 60 liter polyethylene vessel. Samples were taken after elapsed times of 5, 10, 30 and 60 minutes from the start of agitation and were then passed through filter paper Toyoroshi No. 5C (pore size: 1 µ). The concentration of total-Hg in the filtrates was always below 0.5 µg/l.

Mercury Behavior with Pump Circulation

Sixty liters of the mixture was reciprocally moved from one polyethylene vessel (100 liters) to another by a pump. It was then examined to see if the pumping agitation would have any influence on total-Hg contained in the solids. The pump circulation was repeated 10 times and the mixture was allowed to settle for 16 hours. The settled solids were divided into four equal parts (by distance) from top to bottom which were (A) the top layer, (B) the upper layer, (C) the middle layer, and (D) the bottom layer. Total-Hg content was measured for each layer. A mixture that settled for 16 hours without pump circulation was used as a control and total-Hg contents of the settled solids were measured by the same procedure. Figure 8 shows the results in the total-Hg measurement for both the samples. The concentration of total-Hg in the upper layer was higher than in the lower one. Also the concentration of total-Hg in the sample of circulated solids was lower than in the noncirculated sample. Perhaps the fine particles adhering to the larger and heavier ones were detached due to the circulation. It is worthwhile to note here that the mercury in the particles of the sediments are not easily released from the sediment particles to the water, even if they suffer physical disturbance such as agitation or pump circulation.

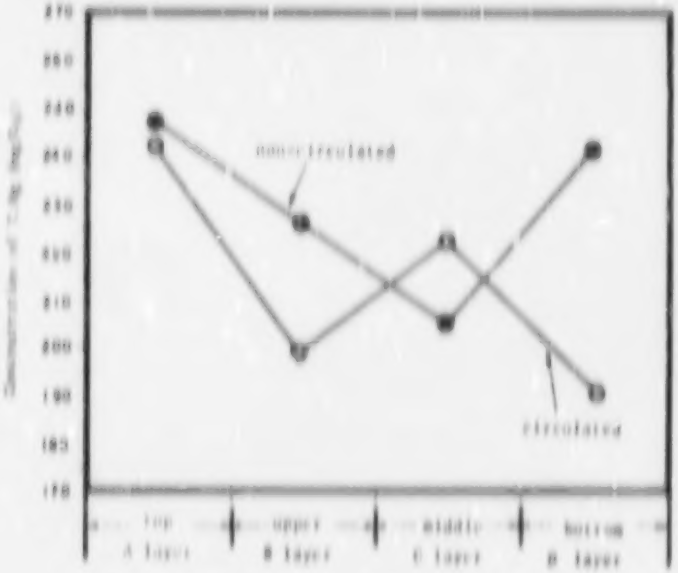


Figure 8. Effects of pump circulation on total-Hg conc.

BEHAVIOR OF MERCURY UNDER FILTRATION

Filtration Test

The supernatent of the settled mixture which was made of 10% (volumetric) sediment was used for filtration tests. Total-Hg and turbidity of this supernatant were 79 mg/kg and 105 degrees, respectively. Three types of filter membranes were employed. They were (1) Membrane Filter RA (Millipore Co. Ltd.); pore size 1.2 ± 0.3 μ; (2) Membrane Filter HA: pore size 0.45 ± 0.2 μ; (3) Membrane Filter VA: pore size 0.05 ± 0.03 μ. Each filtrate of the supernatant through these filter membranes contained a total-Hg concentration of less than 0.5 μg/l.

Permeation Test of Mercury

The tests on the permeability of mercury were carried out in such a way that 3 kinds of embankment fill materials (coal fines, silt, and sand) were packed in columns, and the sediment liquid, diluted by Aquamarine, was supplied under pressure to the tops of the columns.

The test equipment consisted of the 3 filter columns of transparent vinyl chloride, with an inside diameter of 160 mm. They were 1 m, 2 m and 3 m in height. A holding tank of 1 m^3 was employed for delivering pressurized liquid (Figure 9).

The filter media - fine coal, silt and sand - have permeability coefficients of 3.48×10^{-5}, 3.76×10^{-6} and 2.43×10^{-2} cm/sec and mean grain sizes of 19.5, 440 and 490 microns, respectively (Figure 10).

Test conditions were set up as follows:

The inlet pressure at column top	0.5 kg/cm^2
The circulating rate	1.5-1.6 l/min
The concentration of SS	8,460 ppm and 35,000 ppm
The test operation period	15 days cont.

To avoid the accumulation of suspended solids over the surface of the filter bed the liquid was always agitated at the upper part of the columns. The relationships between permeation time and the quantities of permeates are shown in Figure 11.

The permeation rates after 15 days of operation were 0.1-0.15 l/day for coal fines, 0.1-0.3 l/day for silt, and 11.0-21.9 l/day for sand. The permeability of sand was larger by 37-220 times than that of coal fines and silt. The concentrations of total-Hg in the permeates were almost always less than 0.5 μg/l. This is shown in Figure 12.

The relationships between the thickness of the three filter layers and the SS concentration of the permeates are plotted in Figure 13. The effect of thickness on the permeability of SS is remarkable and greatest for the silt material. From these data, it may be concluded that a probability of mercury leakage through the embankment is very small.

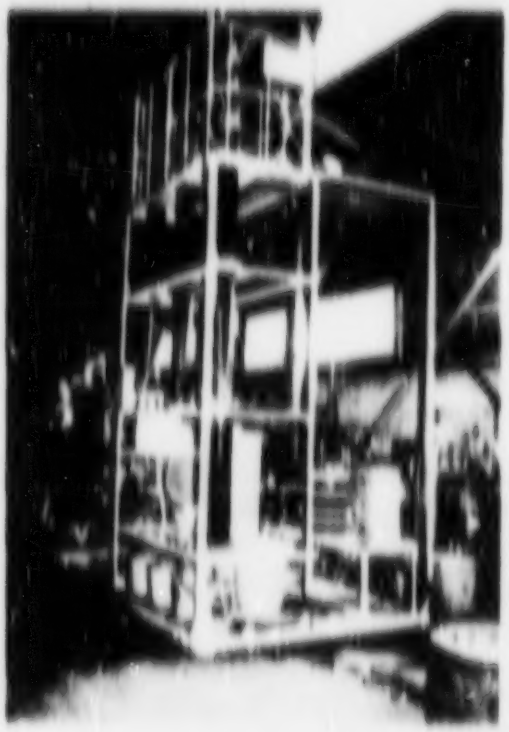


Figure 9. View of permeation test plant.

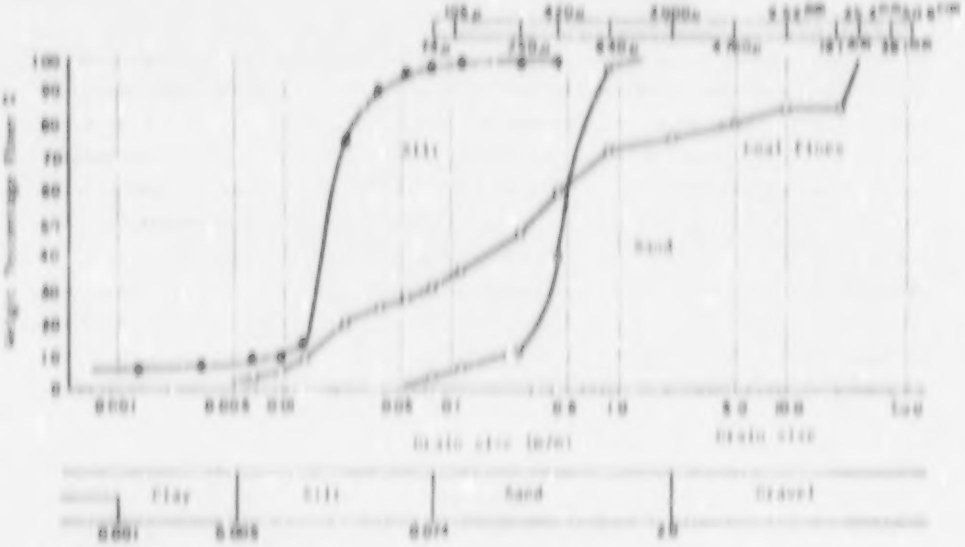
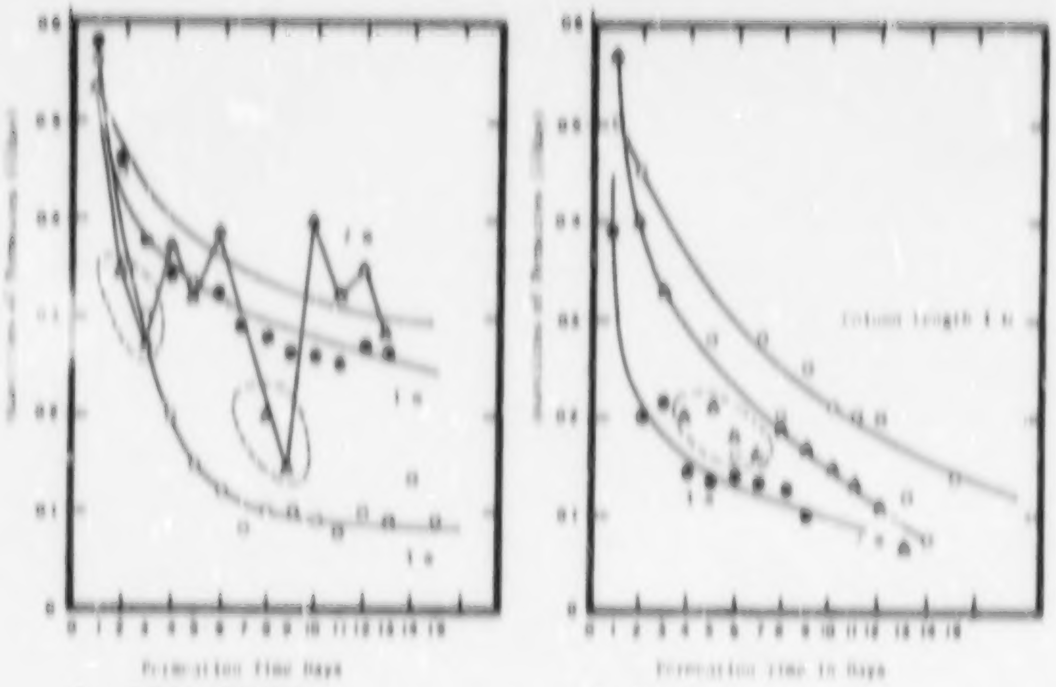
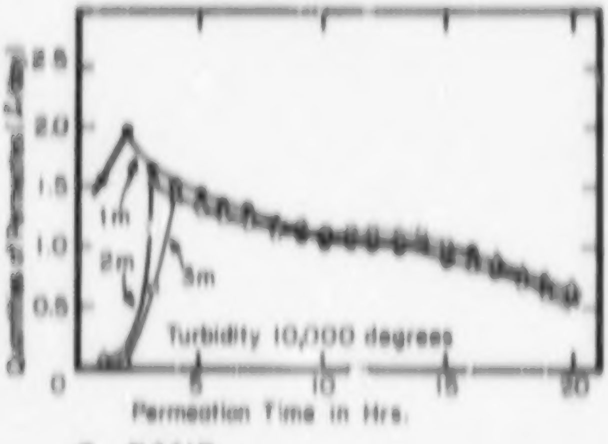


Figure 10. Size distribution of fill materials.



A. COAL FINES

B. SILT



C. SAND

Figure 11. Relationship between permeates and permeation time.

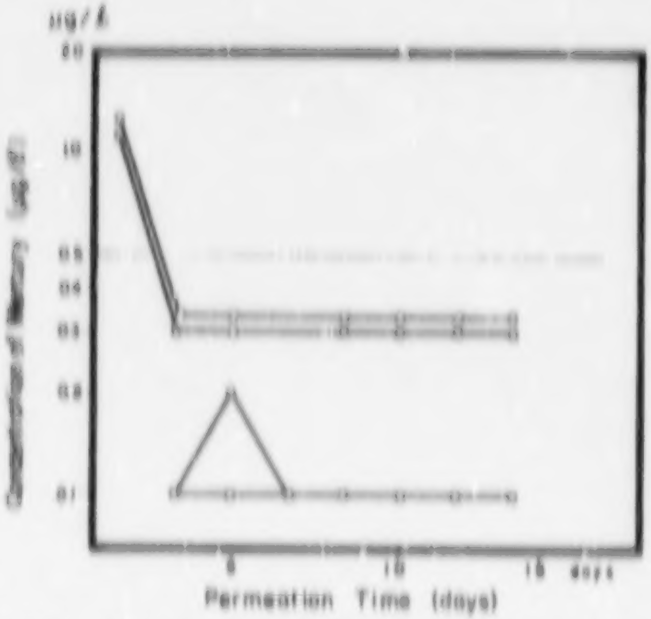


Figure 12. Plot of mercury concentration against permeation time.

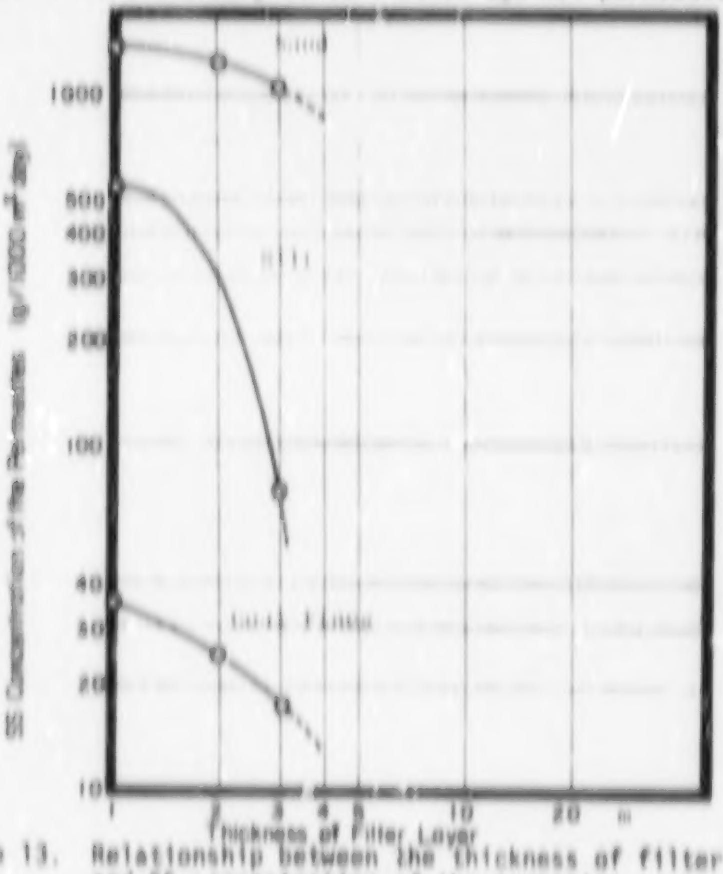


Figure 13. Relationship between the thickness of filter layer and SS concentration of the permeates.

REMOVAL OF SS CONTAINING MERCURY COMPOUNDS

From the preceding test results it has been recognized that mercury compounds are firmly bound to the solid particles of the sediments, i.e., mercury compounds are not easily released by physical disturbances. Therefore, in order to remove mercury compounds, it is necessary to remove suspended solids. It then becomes a matter of importance to estimate the SS-removing capabilities of various sediment treatments.

Coagulation Test of SS Containing Mercury Compounds

Among the various treatments for removing SS, coagulation is the best. As a first step, tests of coagulation were conducted as follows.

- i) The liquid sample of 1 liter is premixed in a beaker (1 liter) for 30 seconds at a mixing rate of 150 rpm.
- ii) A certain amount of inorganic or organic coagulant is added to the beaker and the sample is mixed for one minute at a mixing rate of 150 rpm.
- iii) Then follows a slow mixing at 60 rpm for 3 minutes after which the sample is allowed to settle for 30 minutes.
- iv) To measure various items (flocculating times, sizes of flocs, settling velocities, water properties, etc.) under varying loads of suspended solids five samples were prepared. The SS contents of these samples were 50, 100, 500, 1,000, 5,000 mg/kg.
- v) Two kinds of reagents, inorganic and organic, were used. The inorganic agents, which facilitate clearing, were Alum and PAC. The organic agents, which accelerate precipitation, were sodium alginate, sodium polyacrylate and polyacrylamide. The polyacrylamide is classified into 2 kinds according to the chemical structure, one of them having an electric charge (anion), another non-charged (nonion). The relationship between dissolving time and liquid viscosity of these two kinds of polyacrylamide is shown in Figure 14.

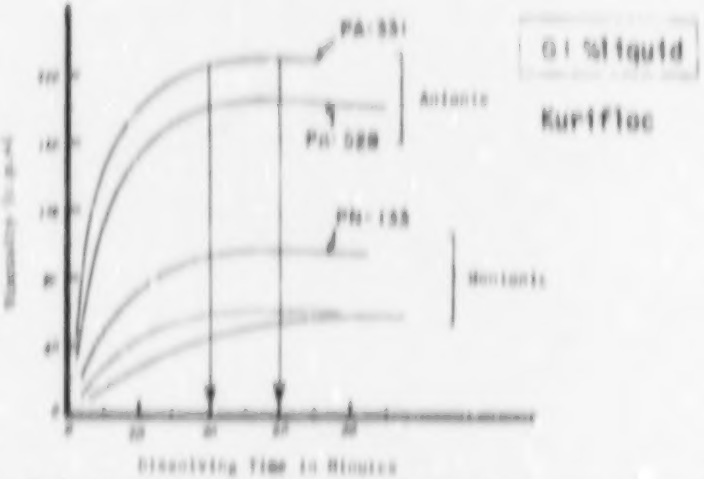


Figure 14. Relationship between dissolving time and viscosity.

Settling by Gravitation

The relationship between the concentrations of mercury and settling times are plotted in Figure 15. The figure indicates that it is very difficult to get the Hg concentration below 0.5 μg/kg in the supernatant, even if a long time is allowed for settling.

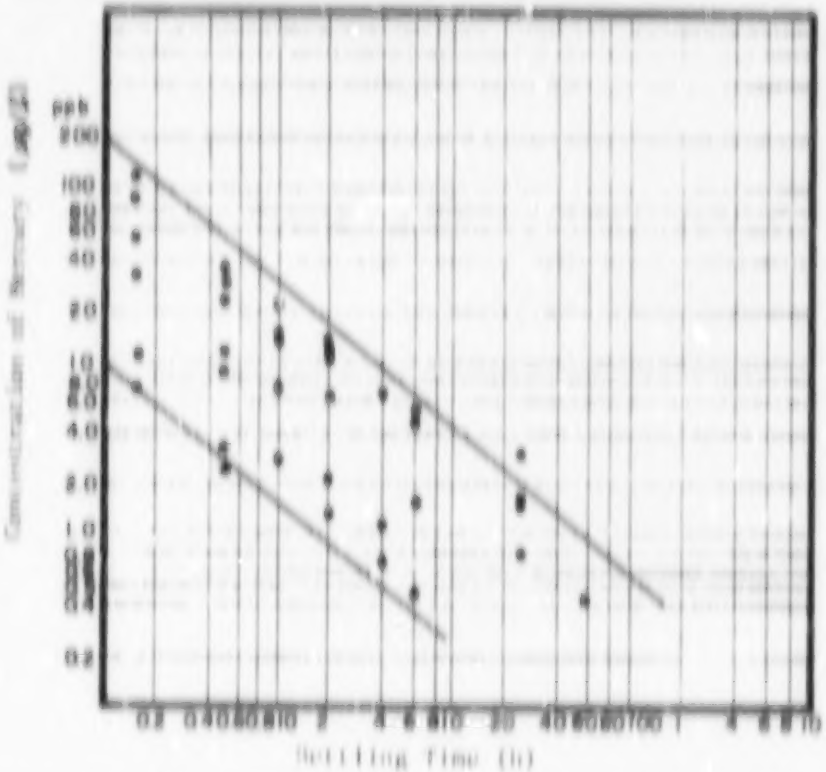


Figure 15. Plot of Hg concentration against settling time.

Settling by Chemical Agents

Figure 16 shows the relationships between the settling velocities of flocs produced and the dosage rates of various kinds of organic flocculants.

From this figure it was found that polyacrylamide was effective in the flocculation of the liquid sample, but both sodium alginate and sodium polyacrylate were not effective. The relationship between the turbidity of the supernatant and the amounts of the flocculant added for the sample having SS of 5,000 mg/l are shown in Figure 17. To obtain a turbidity of 13 to 23 degrees, the required PAC dosage was 100-200 mg/l. It is difficult to get the turbidity below 3 degrees by only using the polymer PA-331. In case of the combined use of PAC and PA-331 a turbidity of below 2 degrees was attained in spite of the small quantities used (only 2-4 mg/l). From this it is clear that the combination of PAC and Kurifloc PA-331 was the most effective coagulant.

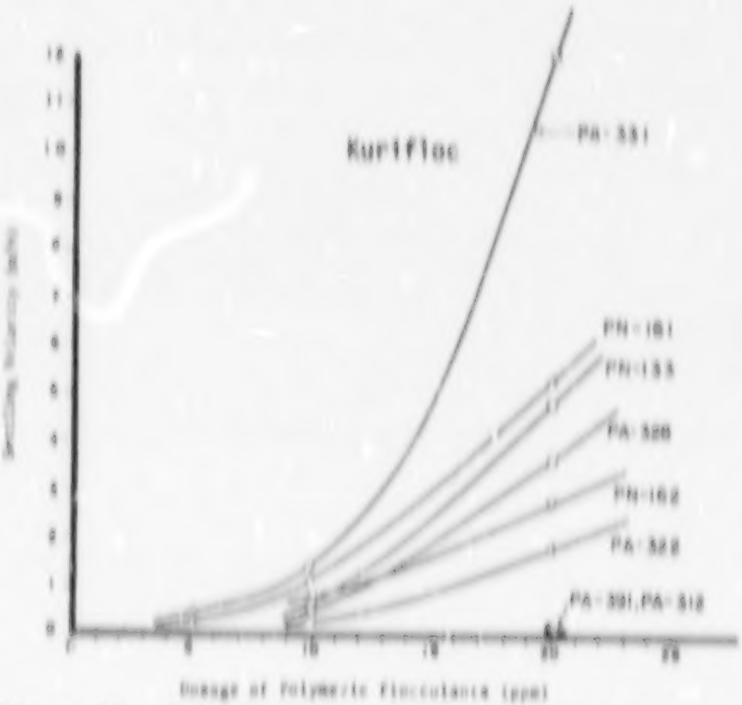


Figure 16. Relationships between settling velocity and dosage of flocculants.

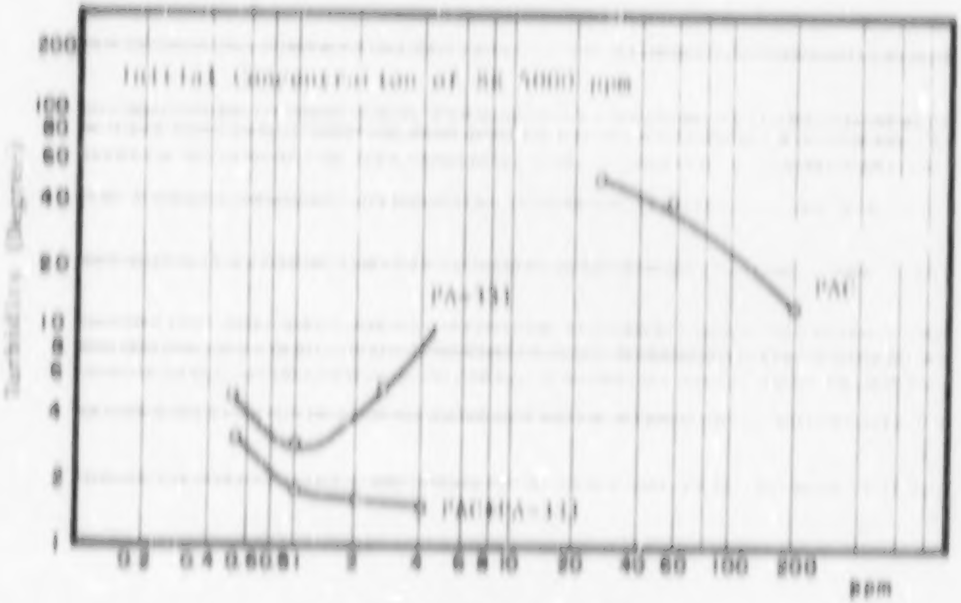


Figure 17. Relationships between turbidity and dusage of flocculants.

Relationships Between the Concentrations of Total-Hg and SS

Figure 18 shows the relationships between the total-Hg concentration and SS remaining in the supernatant. These were obtained by jar-tests of settling with or without flocculants. From this figure, it is clear that the removal of mercury may be achieved by eliminating the suspended solids remaining in the supernatant. Furthermore, it has been proven that in a range of SS around 20 mg/l the total-Hg concentration may be significantly decreased as the SS concentration decreases. The figure also indicates that the reduction to about 10 mg/l of SS in the supernatant may be necessary in order to achieve a total-Hg concentration of less than 0.5 μg/kg.

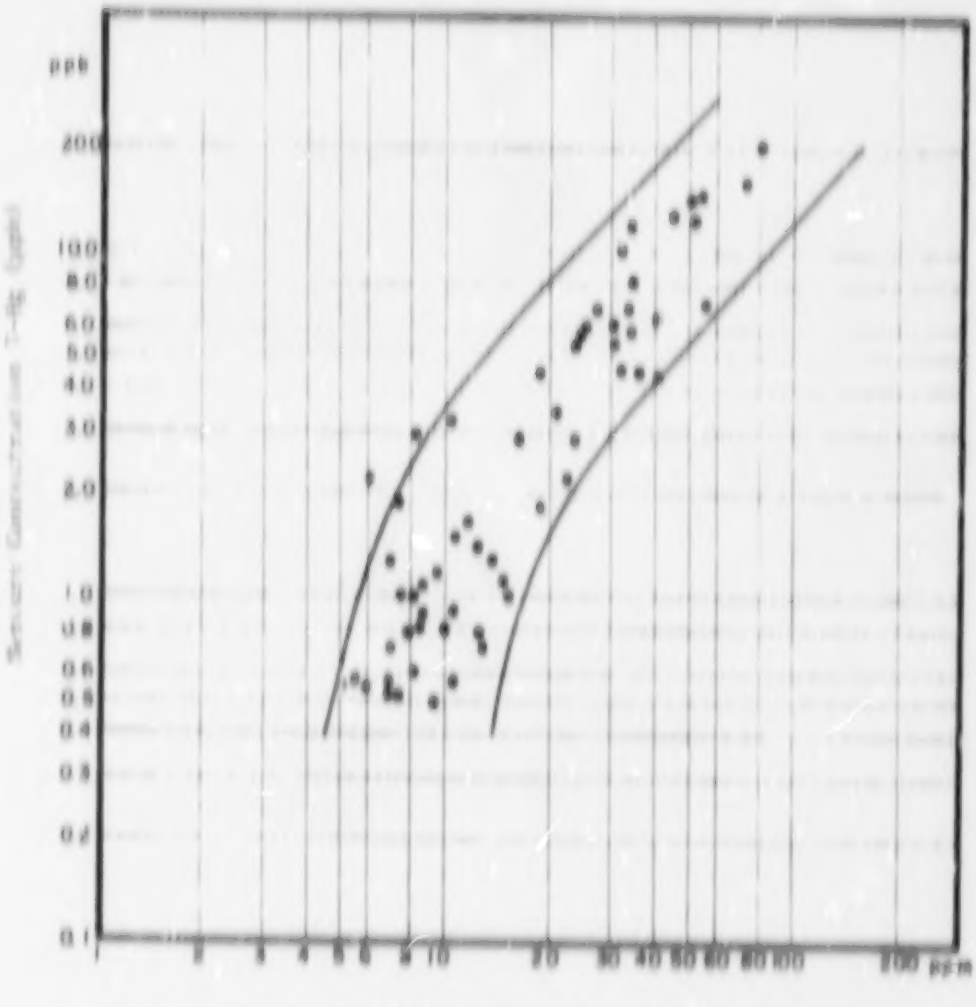


Figure 18. Plot of Hg concentration against SS concentration.

CONCLUSIONS

From the studies the following conclusions have been made.

(1) In order to control the water quality of the final effluent in accordance with "provisional criteria for the removal of bottom sediments," the concentrations of SS in the effluent should be maintained below 10 mg/kg.
(2) In order to obtain a final effluent having an SS concentration of 10 mg/l, it is necessary to treat the spillwater from a settling pond using a clarification plant.
(3) If chemical precipitation is employed to remove suspended solids, the combined use of PAC and polyelectrolite (such as polyacrylamide) is recommended.
(4) In the dredging of the sediments there may be no problem concerning mercury release from dredge spoil particles into the seawater.

USING SAND FILL TO COVER DREDGE SPOILS CONTAINING MERCURY

S. Fujino*
Port and Harbor Bureau
of Kitakyushu City

ABSTRACT

In Dokai Bay of Kitakyushu City, a project to dispose of contaminated bottom sediments containing high concentrations of mercury was planned and carried out. It was the first project to dispose of mercury-containing sediment in Japan. The characteristics of the contaminated bottom sediments and the plan for removal of the sediments were presented and discussed at the first United States-Japan meeting in Corvallis, Oregon in 1975 (1). This paper reports on a project to cover the spoil impounded in a spoil area in order to prevent wind-borne dust from the dried spoil from becoming a secondary pollution source.

To achieve this purpose, a layer of sand one meter thick was placed over the spoil. Since the impounded spoil was too soft to allow the direct placement of the sand layer, the use of a "bamboo-net method" was adopted after studying several alternatives. Rafts made of thick, long bamboos were placed on the mud over the entire spoil area and sheets of plastic were then spread over them. A layer of sand was hydraulically emplaced and followed by a layer of surface soil.

The purpose of the work was achieved safely, securely and economically.

SPOIL AREA

Prior to dredging, a spoil area was prepared by enclosing a basin with a double sheet piled wall. The slopes of the inner sea walls were covered with plastic sheets to keep the polluted water from the spoil from seeping out. The details are in the report presented at the first meeting.

The spoil area was 52,600 square meters and in it impounded some 350,000 cubic meters of polluted material, forming a very soft soil some 7 meters thick at 2.5 meters above the datum level (Figures 1 and 2). The volume of the impounded spoil was almost the same as that of the dredged sediment, i.e., no change in volume due to the dredging operation was observed.

---

* 1-2-7 Nishikaigan, Moji-ku, Kitakyushu-shi, Fukuoka 801, Japan

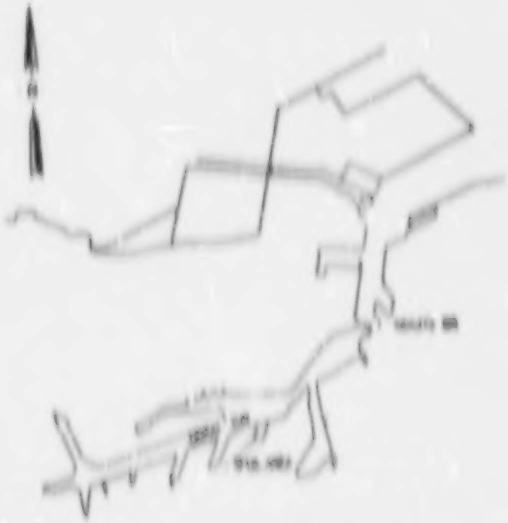


Figure 1. Location of spoil area.

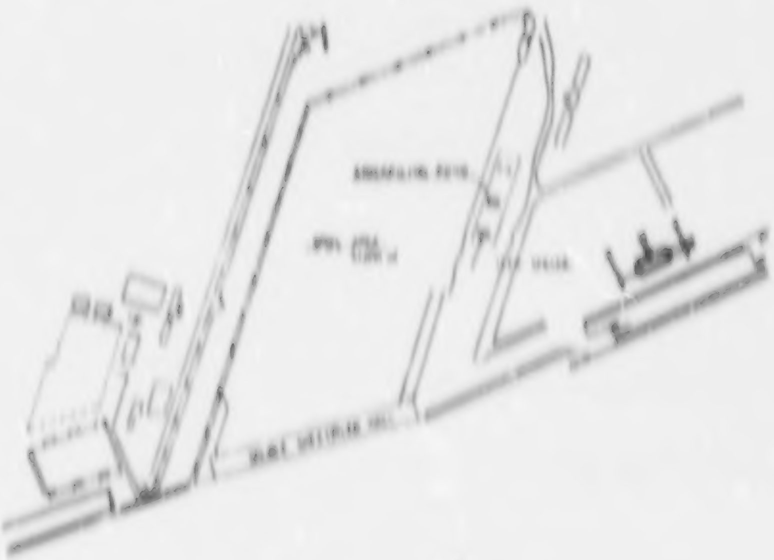


Figure 2. Spoil area.

PROPERTIES OF DREDGED SPOIL

Pollutants contained in the bottom sediments at Dokai Bay were described in the paper presented at the first US-Japan meeting (1). The sediment contained an average of 50 mg/kg of mercury and considerable amounts of lead, chrome, cyanide and arsenic. Alkylmercury was not present. It also contained a significant amount of tar and sulfides--average ignition loss was 16 percent. Mechanical properties of the sediments, obtained from laboratory tests, are as follows:

Cohesion	0.01 kg/cm^2
Compression Index	$Cc = 1.2$
Coefficient of Consolidation	$Cv = 2.5 \times 10^{-2}$ cm^2/min

After the completion of the dredging operation, 85 samples of the spoil were tested and physical properties of the spoil were obtained as follows:

Water Content	63 - 263% (average 128%)	
Specific Gravity of Spoil Particles	2.16 - 2.65 (average 2.44)	
Void Ratio	1.7 - 6.4 (average 3.12)	
Particle Size Distribution	Sand	30% (average)
	Silt	47% (average)
	Clay	23% (average)
Soil Classification	Clayey Silt	

COVERING PLAN FOR SAND FILL

In order to eliminate bad odors and to prevent the dried surface soil from being carried by wind, it was decided that the spoil should be covered as soon as possible.

When the dredging work was complete the spoil was too muddy for any equipment to have access to it. It also contained significant amounts of harmful materials, although they were unlikely to elute from the deposits.

In deciding on a method for covering the spoil area, priority was placed on the worker's safety and health. The covering work also had to be done economically.

The thickness of the covering soil was to be 1.5 meters thick, one meter for preventing subsequent pollution and 0.5 meters to permit vehicle travel.

Five alternative methods were studied to help place the 1.5 meter thick sand fill over the soft spoil impounded in the pond.

a. Covering by means of cloth sheets
Sheets of cloth or woven fabric are often applied to help place a thin layer of sand on soft ground. This method was rejected because the dredged material was extremely soft and any unevenness in the thickness of the sand fill would cause an increase in the volume of sand. Further-

more, eruption of the dredged spoil through the sheets was likely and preventing this is very difficult.

b. Covering by means of sheets and netted ropes
A net made of ropes spread over the above-mentioned sheets has recently been used in Japan. The net is effective in preventing the spoil from erupting, and also makes it possible to bring in earth moving machinery. Unevenness in the thickness of the sand fill was also anticipated and the amount of soil needed was therefore assumed to be great. This method was also rejected.

c. Surface stabilization with stabilizing agents
Instead of covering the spoils with membranes, a portion of the surface layer of a spoil may sometimes be stabilized directly by mixing in a stabilizing agent, consisting mainly of cement, by using floating equipment. But the stabilized layer might be destroyed due to an accidental overload. This alternative also was not considered to be economically feasible.

d. Combined method of b and c
Combined use of the stabilizing agent and a net of ropes was considered very effective and the amount of sand fill could be reduced compared to method a or b. This method, however, was very expensive and was accordingly eliminated.

e. Covering by means of sheets and rafts of bamboos (Bamboo-net method)
In this method rafts of bamboos are placed on top of the mud and then sheets of cloth are spread over them. Since bamboos are resilient the rafts will distribute the load of sand fill equally on the soft spoil and will reduce the needed volume of sand to a minimum. The rafts will enable workers to lay the membrane safely since the rafts are used as footholds. This method was finally chosen for its economy and safety.

Two alternatives were studied in placing a thin sand layer over the bamboo rafts and the sheets of cloth. One used a conveyor system and the other used a small hydraulic pump dredge to transport a mixture of sand and water.

The former was rejected because it would be more costly and take more time. The latter was finally selected for its economy and its certainty in producing an even, thin layer of sand.

BAMBOO RAFTS

Bamboo rafts were used to distribute the weight of sand placed over them. In order to determine the spacing of the bamboo members an elastic foundation analysis was performed. Although the ground consisting of the dredged spoil was not elastic, the analysis was performed since there was little experience with this kind of work.

A bamboo is known to have a Young's modulur of 120,000 kg/cm^2 and a tensile and compressive strength of 800 kg/cm^2. An eight meter long bamboo with a diameter of 5 cm and a thickness of 0.5 cm has a section modulus of 7.245 cm^3 and a maximum resisting bending moment of 5,796 kg/cm.

Assuming that bamboos are beams spaced about 70 cm apart on an elastic foundation the elastic modulus of which is 0.03 kg/cm^3, and also assuming that the first layer of a fill be placed at one time as shown in Figure 3, the bending moment, stress and vertical deflection were computed as follows:

Bending Moment	4,660 kg/cm
Stress	450 kg/cm^2 (Factor of Safety 1.78)
Vertical Deflection	19 cm

From this analysis it was expected that a raft of bamboos spaced 70 cm apart both laterally and longitudinally could withstand the weight of a layer of sand laid as shown in Figure 3. It was decided that the one meter thick sand fill was to be placed by repeating the operation three times.

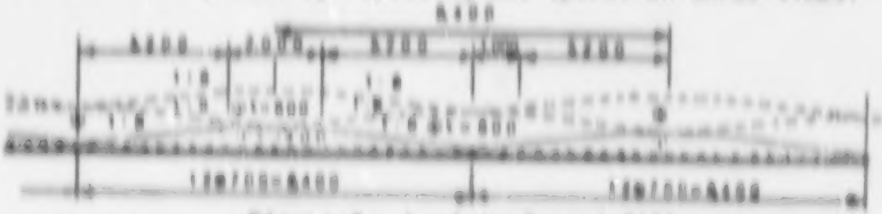


Figure 3. Laying of sand fill.

LAYING OF BAMBOO RAFTS

Prior to the laying of bamboo rafts the surface water of the dredged spoil was pumped into a settling pond where a high-molecular flocculant was added. The 8 m x 8 m bamboo rafts were prefabricated outside the spoil area and spread over the spoil by laborers with the assistance of a truck crane and winches (Figure 4).

Figure 4. Bamboo raft construction.

To protect the workers from accidents, life jackets, rubber gloves and rubber boots were supplied to all the workers, who always worked in pairs. A number of boards for footing were used to prevent the workers from falling. A guard was posted to ensure the safety of the workers. A portable shower house was on site for use when the workers got soiled or fell into the spoil.

SHEETS

The main purposes of the sheets were to prevent the spoil from erupting and to prevent the fill material from collapsing into the mud during the filling operation. The sheets also served as footholds for subsequent work.

To determine the type of membrane, a calculation was carried out which showed that the sheets should have a tensile strength not less than 16 kg per centimeter of width. A polypropylene cloth was finally chosen. Its properties are listed in Table 1. For safety, spaced polypropylene ropes were laid between each bamboo and fastened to the bamboo rafts (Figure 5).

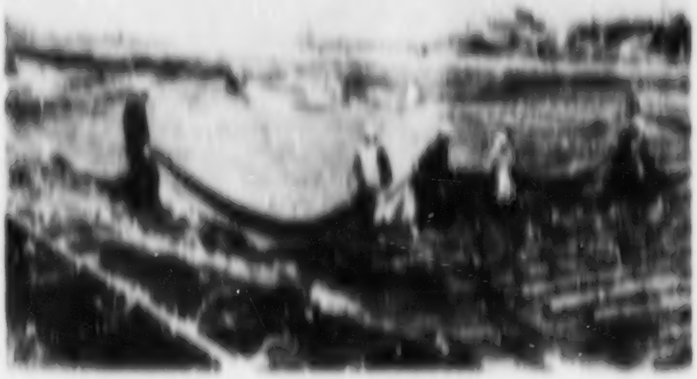


Figure 5. Illustration of safety techniques.

TABLE 1. SPECIFICATIONS OF POLYPROPYLENE COVER SHEETS

Material	Polypropylene PP-1616	
Number of mesh	longitudinal lateral	16/inch 16/inch
Tensile strength	longitudinal lateral	103.3 kg/5 cm 100.0 kg/5 cm
Elongation	longitudinal lateral	25.1% 14.8%
Tensile strength of sewed portion	97.2 kg/5cm	

42 m x 22 m polypropylene membranes were manufactured at the factory and delivered to the site. They were joined together after being spread over the bamboo rafts. The detail of the joint is shown in Figure 6.

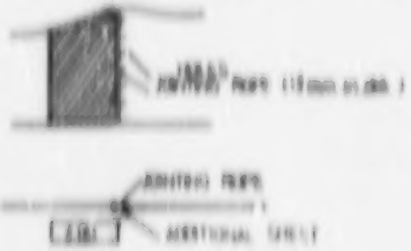


Figure 6. Joint of sheet.

PLACEMENT OF SAND FILL

Clean sea sand was collected by sand carriers equipped with grab-buckets and transported to a nearby concrete wharf where the sand was unloaded, transported to the site by dump trucks, and stockpiled in a pond by means of belt conveyors. The pond was 85 m long, 18.6 m wide and 3 m deep. The grain size distribution of the sand was as follows:

D max. = 10 mm $\quad$ D_{60} = 0.47 mm
D_{30} = 0.29 mm $\quad$ D_{10} = 0.18 mm

Sea water was let into the stockpiling pond and three portable hydraulic pump dredges were used to move the sand-water mixture to the fill area. The dimensions and capacity of the dredges are as follows:

Length	8 m
Width	2.5 m
Depth	0.8 m
Draft	0.6 m
Main Pump	Diesel Engine 165 - 175 HP
Diameter of Discharge Line	8 inches
Maximum Length of Pipe Line	600 m
Maximum Discharge	300 cu. m per hour (water) 60 cu. m per hour (sand)

The dredges sent the sand-water mixture through discharge lines. Each outlet of the discharge line was extended at a rate of 4 meters every five minutes, placing the sand fill as shown in Figure 3. The water was recovered and pumped back into the stockpile pond to be reused since it contained no pollutants according to laboratory analyses.

The quantity of sand necessary to provide a minimum thickness of 1 meter was calculated as follows: the difference between the largest and the smallest displacement of the sheet surface due to elastic compression and plastic flow of the spoil was assumed to be 30 cm. The difference in further displacement due to uneven consolidation was assumed to be an additional 30 cm.

Since the surface of the completed ground had to be flat, the mean thickness of the sand fill was assumed to be 1.3 meters (Figure 7).

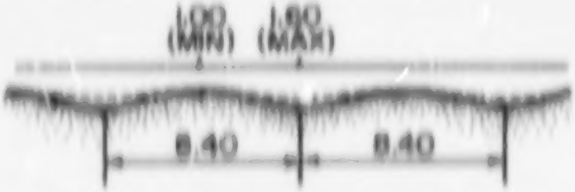


Figure 7. Assumed formation of sand.

The volume of sand to be transported was increased by an additional 15% to account for volume changes due to the effect of compacting during placement and loss during handling. After the sand laying was completed the recycled water was purified in the settling pond and discharged to the sea. Numerous soundings were made and they showed that the required minimum thickness was obtained with a mean thickness of 1.31 meters.

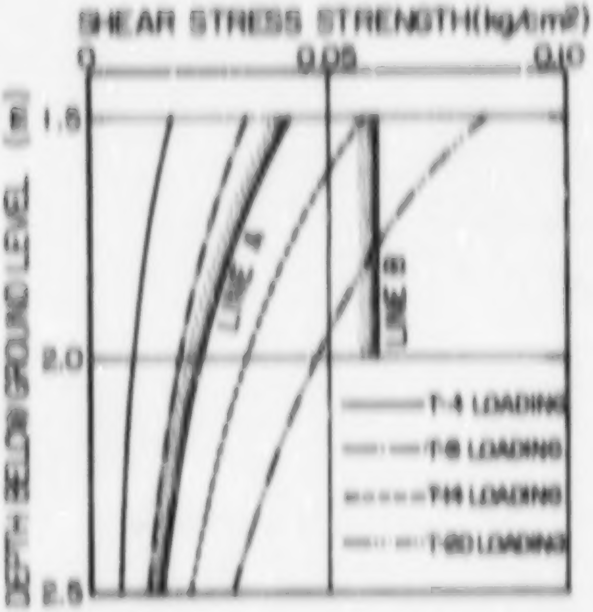
PLACEMENT OF SURFACE SOIL

Two months after the sand fill was completed placement of surface soil was begun to prevent windblown sand dust and to increase the ability of the spoil area to support traffic. Two months following the sand placement the covered spoil had been consolidated to such a degree due to the weight of the sand that small earthmoving equipment could be operated on it. Figure 8 shows the shear stresses caused in subsurface soil by four different truck loads. Line A in Figure 8 shows the cohesion of the subsurface silt 60 days after the sand placement. It indicates that a 4 ton dump truck (total weight appros. 8 metric tons) or lighter equipment can travel on the sand. Three-ton bulldozers were used to place 20 cm thick finger dikes as shown in Figure 9. These acted as counterweights. They were 30 cm lower than the final level of the ground. The interspaces between these dikes were then filled, bringing them to the same level as the dikes, after which a 30 cm thick layer of soil was laid over the whole area using 12-ton bulldozers. The 3-ton bulldozers remained at least 20 meters ahead of the follow-up bulldozers, i.e., the first stage of the dikes extended at least 20 meters in front of the completed area.

RESULTS AND DISCUSSION

Main materials used in the fill and cover project are listed below:

Bamboo Rafts	52,600 sq. m	
Bamboos, 5 cm in diameter, 8 m long		30,700 pcs.
Annealed iron wire, 3.5 mm in diameter		11 tons
Polypropylene ropes, 12 mm in diameter		162,000 m
Membrane	52,600 sq. m	
Polypropylene woven cloth, PP 1616, 42 m x 22 m		82 sheets
Sea Sand	82,000 cu. m	
Surface Soil	33,000 cu. m	



LINE A:
Estimated cohesive strength
at 60 days after completion
of sand fill.

LINE B:
Estimated final cohesion

Figure 8. Subsurface stress by a dump truck.

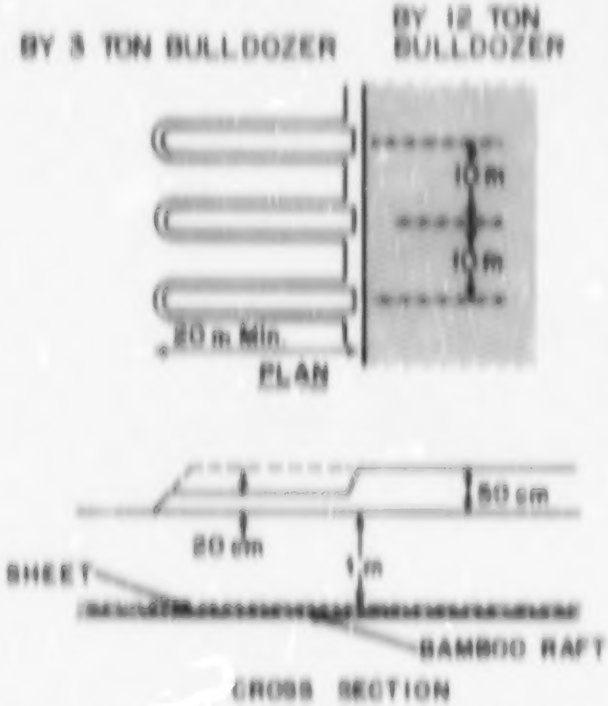

BY 3 TON BULLDOZER
BY 12 TON BULLDOZER
10 m
10 m
20 m Min.
PLAN
50 cm
20 cm
1 m
SHEET
BAMBOO RAFT
CROSS SECTION

Figure 9. Procedure of surface soil placement.

Settling due to consolidation is being measured. Sixty cm of settlement was recorded four months after the completion of the entire project. In order to detect the level of contamination of the covering sand due to effects of filtration and adherence, twelve samples were taken at six points. One sample was taken from the upper part and one from the lower part of the sand layer at each point. Samples were analyzed at the Kitakyushu Municipal Institute of Environmental Health Sciences. The concentrations of main contaminants in the sand are shown in Table 2. It shows that the sand at the level 30 cm above the sheet was not affected to a great extent by the contaminants contained in the spoils. In another laboratory experiment (S. Yamada, et al., 1975) similar results were reported (2).

> "In this experiment a sand layer of 50 cm in thickness was placed on a polluted mud filled in polypropylene bags to squeeze its pore water out of the mud. The concentration of suspended solids in the water taken from the sand 30 cm above the bag surface was 10 parts per million. Visual examination showed that the mud which seeped out of the bags remained in the area less than 10 cm above the bag surface." (sic)

TABLE 2. CONCENTRATION OF CONTAMINANTS IN THE UPPER AND LOWER PORTION OF THE SAND LAYER COVERING THE TOXIC SPOILS

Contaminants	Concentration (mg/kg)		
	In stockpile pond	Upper[a] part	Lower[b] part
Cadmium and its compounds	0.033	0.05	0.06
Cyanides	--[c]	--	--
Organic phosphorus	--	--	--
Lead and its compounds	2.2	2.2	2.6
Hexachrome	--	--	--
Arsenic and its compounds	--	--	--
Mercury and its compounds	0.014	0.008	0.012

[a] Upper part: 70 cm above the sheet [b] Lower part: 30 cm above the sheet

[c] --: "not detected"

Most of the pore water squeezed out of the spoil will rise through the covering soil and evaporate. Some of it might flow horizontally through the sand fill into surrounding areas. It was unlikely that pollutants would move to surrounding areas since most of the pollutants are bound to suspended solids which would be retained in the sand. To test this hypothesis several wells were installed in the periphery of the spoil area to monitor the quality of ground water. Monitoring of the wells has continued and has shown no abnormalities to date.

CONCLUSIONS

The "bamboo-net method," introduced as a surface treatment technique to confine toxic spoils which do not elute, was verified as a satisfactory technique from the following points of view:

1. The technique does not allow secondary pollution.
2. It is safe and sanitary for the laborers concerned.
3. It is economically feasible.

It was also found that a covering soil of 100 cm in thickness was sufficient in itself to prevent secondary pollution (such as dust and bad odors) from occurring. There was also no influence on the periphery of the area due to exudation of pore water from the spoils. Execution of the technique does not require sophisticated skills. The proposed thickness of the covering soil was obtained within a reasonable period of time by applying the technique to the spoil surface step by step. This allowed the use of the surface by vehicular traffic in a reasonable time.

The authors hope that this paper will be of some help to those who are concerned about the management of polluted bottom sediments.

REFERENCES

1) T. Koike. Real application of management techniques in Port Kitakyushu, The US-Japan Experts Meeting on Management of Bottom Sediments Containing Toxic Substances, November 1975.
2) S. Yamada, K. Mori, N. Morii. Experiment on disposal of toxic mud by confining in bags of cloth, June 1975.

CHEMICAL STABILIZATION OF SOFT SOILS

T. Okumura*
Chief, Soil Stabilization Laboratory
Port and Harbor Research Institute
Ministry of Transport

ABSTRACT

Chemical stabilization methods have recently been employed on reclaimed surface soils and in-situ soft clayey soils. Chemical stabilization is affected by the characteristics of the soil and the stabilizing agents. In this paper various kinds of chemical stabilizing agents are reviewed, stabilizing effects are considered, and the test results of comparisons of different stabilizers are shown. In conclusion, there is no single agent useful for all kinds of soil and stabilizing conditions. In many cases it is more reasonable to use a slightly greater amount of a simple agent than to use expensive additives.

INTRODUCTION

Toxic sediments have often been treated by means of reclamation. In these cases the surface should be stabilized as soon as possible to reduce the possibility of secondary pollution and/or to utilize the reclaimed land.

Recently, chemical stabilization methods have been employed in these cases as well as physical or mechanical methods such as sand spread and covered with sheets. Chemical stabilizations are also employed in improvement of in-situ soft soils (Okumura and Terashi, 1975).

In general the principle of chemical stabilization utilizes the chemical bonding of soil and stabilizer when they are mixed together. However, the stabilizing effect of chemical agents also depends on many factors such as water content, grain size, physio-chemical properties of soil and the type of agent. For example, Figure 1 shows the variety of stabilizing effects of quicklime on various chemical agents.

CHEMICAL STABILIZING AGENTS USED IN PRACTICE

Table 1 summarizes the chemical stabilizing agents commonly used on the reclaimed surface layer and on the in-situ soft soils.

---

* 3-1-1 Nagase, Yokosuka-shi, Kanagawa 239, Japan

TABLE 1. CHEMICAL STABILIZING AGENTS USED IN RECLAIMED SURFACE AND IN-SITU SOFT SOIL

Type	Main Component	Name of Agent	Phase in Use	Method of Treatment	Name of Treatment
Lime		Quicklime	solid	mixing (deep/shallow)	Deep-Lime-Mixing, etc.
	Quicklime	Chemico Lime -300	solid	mixing (shallow)	Chemico Lizer
				lime pile	Chemico Pile
		[illegible] Lime	solid	mixing/lime pile	
	hydrated lime	hydrated lime	solid/slurry	mixing (shallow/deep)	
		Road Lime	solid	mixing	Soil Liner
	slag with lime	Slag with Lime	solid	mixing	
		Slag [illegible]	solid	mixing	slag, gypsum and hydrated lime
	gypsum and lime	Gypsum with Lime	solid	mixing	
	sludge ash and hydrated lime	[illegible]	solid	mixing	
Portland Cement	Portland Cement	Portland Cement	slurry/solid	mixing (deep/shallow)	Deep Chemical Mixing, etc.
		Cement Mortar	slurry	mixing (deep)	Clay Mixing Consolidation
		Fuji Beton-PC	slurry	mixing (shallow)	[illegible] Method
		Fuji Beton [illegible] -PC, -PL, -AL	slurry	mixing (shallow)	
		[illegible] Mix	slurry	mixing (shallow)	Consolidator System
			slurry	sludge mixing	[illegible] System
	Portland Cement and hydrated lime	Chemico Lime -200, -400	slurry	mixing (shallow)	[illegible], M.R.
			solid	mixing (shallow)	Mixer
	Portland Cement and slag	Slag Cement	slurry	mixing (shallow/deep)	
asphalt				mixing with soil	
additive		Denka [illegible]-1, -2		with cement, for rapid solidification	
		[illegible]		ditto, for Hg fixing	
	polymer	[illegible]		ditto, for fixing metallic compounds	
	polymer	[illegible] 35		ditto, for making impervious film	
		[illegible]		ditto, for organic soils	

Lime and its Variations

Quicklime

(1) Quicklime

Quicklime is one of the most popular and widely used agents in road construction for sub-base and sub-grade stabilization. Recently, the Deep-Lime-Mixing Method was developed (Ōkumura, et al., 1974) and has been used for stabilization of the in-situ soft clayey layer in both marine and terrestrial projects. In this case high activity quicklime is effective because of its quick hydration and ease of mixing.

(2) Chemico Lime - 100

Chemico Lime-100 is quicklime with some additives and is produced by Onoda Cement Co. Ltd. It is used for both road construction as Chemico Lizer and for deep stabilization by means of lime piles as Chemico Pile.

(3) Semii Lime

Semii Lime consists mainly of quick lime and is used by Fujita Industry Co. Ltd. for road construction or as lime piles.

Hydrated Lime

(1) Hydrated Lime

Hydrated lime is used as widely for road construction as quicklime. It is possible to use it in the form of slurry for deep mixing stabilization.

(2) Road Lime

Road Lime consists mainly of hydrated lime with a small amount of water. It is used by Nisshin Hōdō Co. Ltd. for road construction.

Other Agents with Lime

(1) Slag and Lime

Slag (blast furnace slag) is not very effective in the pure state for rapid soil stabilization. But with the aid of quick or hydrated lime, its reactivity becomes greater and more rapid. In some cases the mixture of slag and lime is more effective than pure lime. Several steel companies and others have studied the effectiveness of the slag and lime mixtures.

(2) Slag Bacillus

Slag Bacillus consists mainly of slag and contains gypsum, hydrated lime and other materials. In some cases, it is more effective than quicklime (Kotani, et al., 1974).

(3) Gypsum and Lime

Gypsum in its pure state has almost no effect for soil stabilization. But, with the aid of lime, it may be more effective than pure lime.

(4) Hi-Tog

Hi-Tog is sludge ash from sewage with some hydrated lime. It is produced by Hirose Steel Industry Co. Ltd. In some cases it is more effective than pure hydrated lime. It also allows utilization of a waste product.

Portland Cement and its Variations

Portland Cement

(1) Portland Cement

Portland cement has been widely used in road construction as a "soil cement" for solid states. Recently it has been used for solidifying soft dredged soils (e.g., the Takenaka Sludge Treatment System), and also has been tried mixed with deep in-situ clayey soils in the liquid state as Deep Chemical Mixing Method or DCMM method.

(2) Cement Mortar

In some cases, cement mortar (a slurry of cement and sand) is a more effective stabilizer than cement slurry. It has been used for in-situ soil stabilization as the Clay Mixing Consolidation method by Fudo Construction Co. Ltd.

(3) Fuji Beton

Fuji Beton - FPC and Fuji Beton Toa -PC, -FK, -AL are compounds consisting of portland cement with various additives (organic and/or inorganic). They have been widely used for sludge treatment and for surface stabilization of reclaimed land.

(4) Fudo Mix

Fudo Mix is a series of portland cement compounds with various kinds of additives, and is used for surface treatment as the Consolider System.

Portland Cement with Other Agents

(1) Chemico Lime -200, -400

Chemico Lime -200 and -400 are portland cement with some amount of hydrated lime. They are used in liquid state for surface treatment as to Mini-max method or M.R. method, and as the Mixer method in the solid state.

(2) Slag Cement (Portland Blast Furnace Slag Cement)

Slag cement is, in some cases, more effective and economical than portland cement. It is also widely used in surface and deep stabilizations.

Asphalt

Asphalt is also used in sludge treatment, but it is not as popular as lime or portland cement.

Additives

There are several additives for portland cement which improve its stabilizing qualities. Denka HS-1 and -2 are added for rapid solidification. Konseal makes films on the surface of organic soil grains in the same way as Danseal X5. To fix metallic compounds Nisso ALM and EB-CX are employed.

GENERAL EFFECTS OF STABILIZATION WITH QUICKLIME

Variations which Depend on Soil Type

As shown in Figure 1, the unconfined compressive strengths, q_u, obtained from laboratory mold tests in accordance with the authors' manual (PHRI, 1975), very widely depending on the kind of soil. Strength increase due to consolidation, being a mechanical stabilization, does not depend as much on the kind of soil. The c_u/p ranges from 1/3 to 1/4. Thus, the reactivity of lime treated soil depends mainly on the chemical and mineralogical properties of the soils, and therefore has a variation greater than 5 times that shown in Figure 1. In general the effectiveness of lime or cement on organic soils is less.

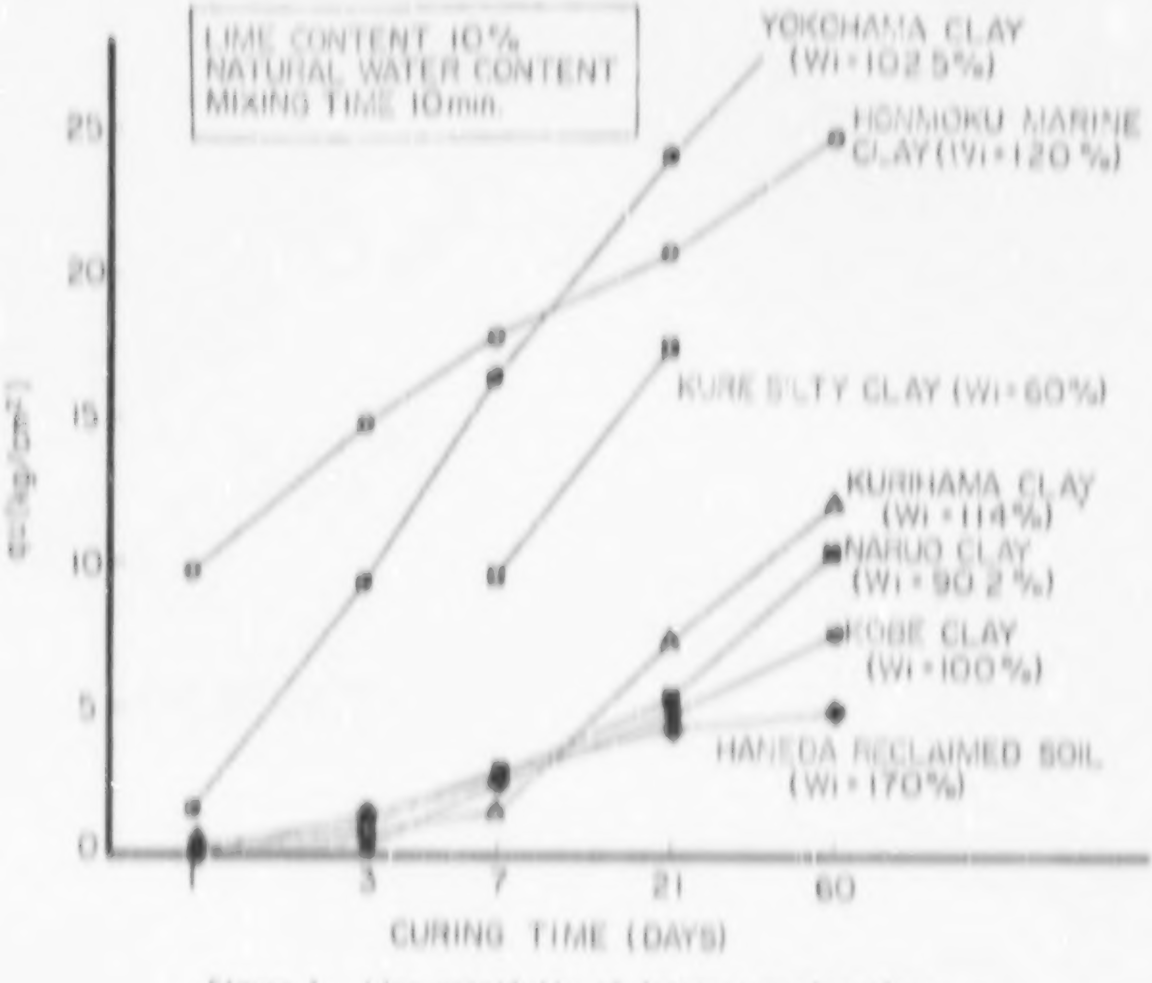


Figure 1. Lime reactivity of Japanese marine clays.

Effect of Time and Temperature

Strength increase with time, of lime treated soil, is similar to that of cement concrete and as shown in Figure 1, is roughly proportional to the logarithm of time. High temperature results in a more rapid reaction between the soil and the lime leading to a quick increase in strength. The final strength reached does not necessarily depend on the temperature.

Effect of Initial Water Content

Figure 2 shows the effect of the initial water content of the treated soil on the lime reactivity. The quicklime content relative to the solid weight of the soil (%) is represented as a_w, and T_c is the curing time (days). The reactivity is maximum at an initial content near the "liquid limit," w_L, or a little less. For higher water content the reactivity is greater than that fur lower values of water content, and therefore the effectiveness for stabilizing dredged material with high water content may still be considerable.

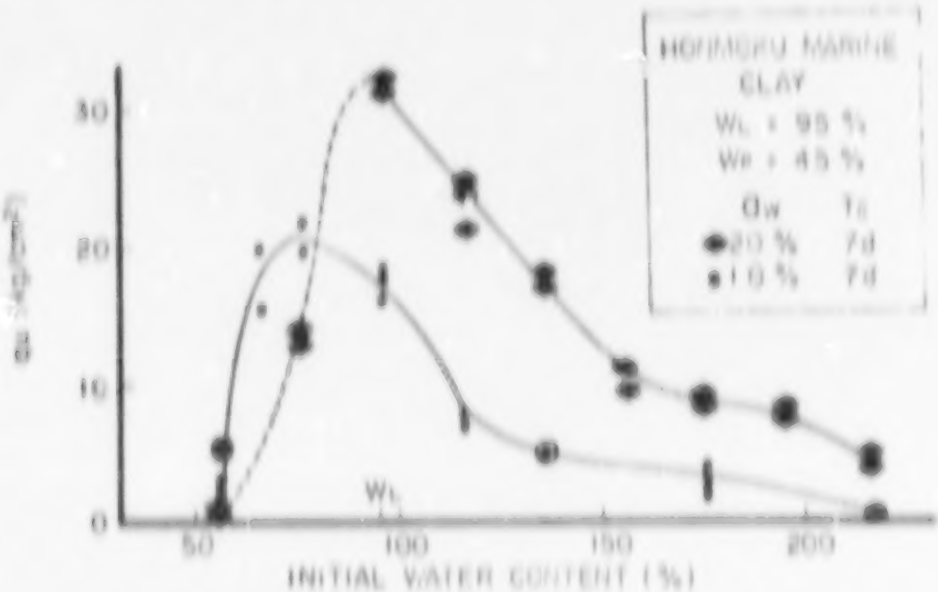


Figure 2. Changes in lime reactivity with initial water content.

Effect of Lime Content

The lime reactivity is roughly proportional to the lime content for small amounts of lime. However, it is not necessarily true that more lime results in more reactivity. In the extreme case 100% quicklime becomes 100% hydrated lime, which is a rather soft material. In the practical range of lime content, which is up to 15 or 20%, this trend of diminishing effectiveness sometimes exists.

Effect of Grain Size

Contrary to common sense, lime reactivity is not maximum in pure clay, but is higher in soils with some sand content (Yanase, 1968). For pure sand the lime reactivity is less.

Consolidation Characteristics of Lime Treated Soils

Figure 3 shows an example of void ratio, e, vs. consolidation pressure, p, for a soil-lime mixture cured for 3 weeks. The compressability of treated soil is much less than that of untreated soil up to the consolidation yield stress, p_y. It means that the lime treated soil is more resistant to settling than the untreated. The consolidation yield stress or the preconsolidation pressure of the treated soil is proportional to the unconfined compressive strength, q_u, as shown in Figure 4. Therefore, the greater the strength increase, the heavier the allowable load is before settling begins.

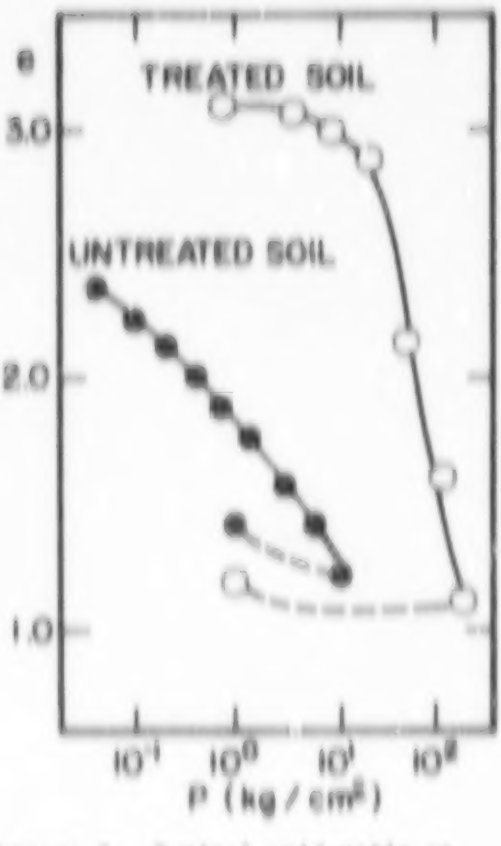


Figure 3. Typical void ratio vs consolidation pressure curve.

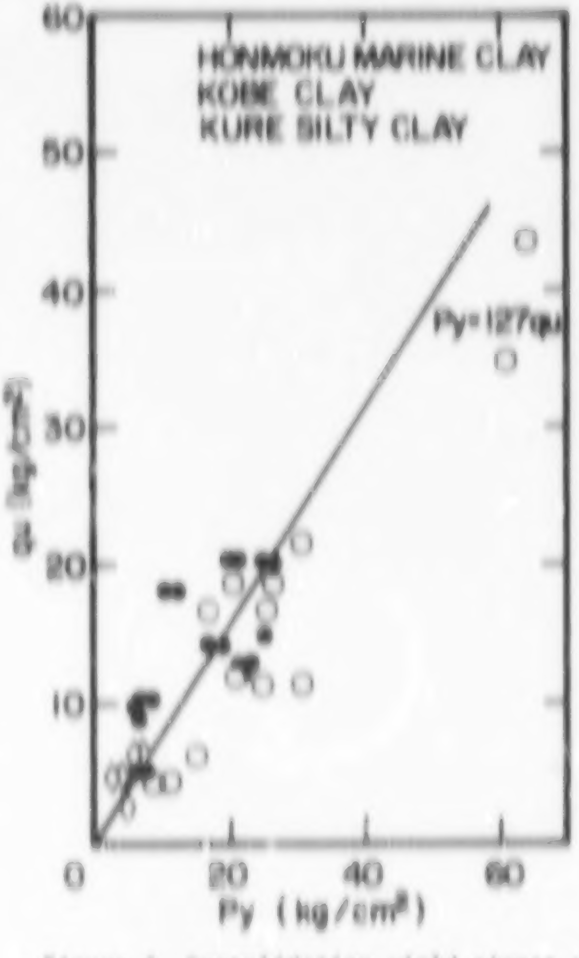


Figure 4. Consolidation yield stress vs unconfined compressive strength.

Figure 5 shows a relationship between the ratio of the coefficient of volume compressability of treated soils, m_v, to that of remolded and untreated soils, m_{vr}, and the mean relative consolidation pressure to the yield stress, p/p_y. The relative compressability, m_v/m_{vr}, is less than 10% for a mean relative consolidation pressure less than 0.3, and is about 1.5 for an m_{vr} greater than 1.0.

Relationship between the relative coefficient of consolidation, c_v/c_{vr}, and the mean relative consolidation pressure is shown in Figure 6. The rate of consolidation of treated soils is more than 10 times greater when compared with that of the remolded and untreated soils in the range of relative consolidation pressures less than 1.0 (Okumura, Terashi and Yoshida, 1974).

COMPARISON OF THE STABILIZING EFFECTS OF VARIOUS CHEMICAL AGENTS ON SOME SOFT CLAYEY SOILS

The author and his colleagues carried out a series of laboratory tests to compare stabilizing effects of various chemical agents. The testing method was in accordance with the author's manual (PHRI, 1975), and the stabilizing effect is evaluated by employing the unconfined compression test.

Physical and chemical properties of the soils used in the test are listed in Table 2. Two of the soils are organic and the other two are inorganic. Chemical agents mixed with these soils are two of the quicklime type, two of the portland cement type, plus three additives used with the portland cement.

TABLE 2 PHYSICAL AND CHEMICAL PROPERTIES OF TESTED SOILS

Property	Soil			
	K	M	T	Y
Specific gravity of solid, G_s	2.66	2.71	2.44	2.52
Liquid limit, w_L, %	91.8	89.9	145.4	137.0
Plastic limit, w_p, %	40.9	27.9	48.2	55.8
Soil classification	clay	silty clay	sandy silt	clay
pH	8.1	8.4	7.8	8.1
Ignition loss, %	7.9	9.7	19.2	20.0
Organic carbon, mg/g	10.5	8.5	29.5	23.5
Sulfide, S mg/1 g solid	0.26	0.67	3.31	19.16
Total Hg, mg/kg		0.36		4.66
Oil content, mg/kg	190	2120	14500	19400
COD, O_2 mg/1 g solid	10.0	16.7	46.7	86.3

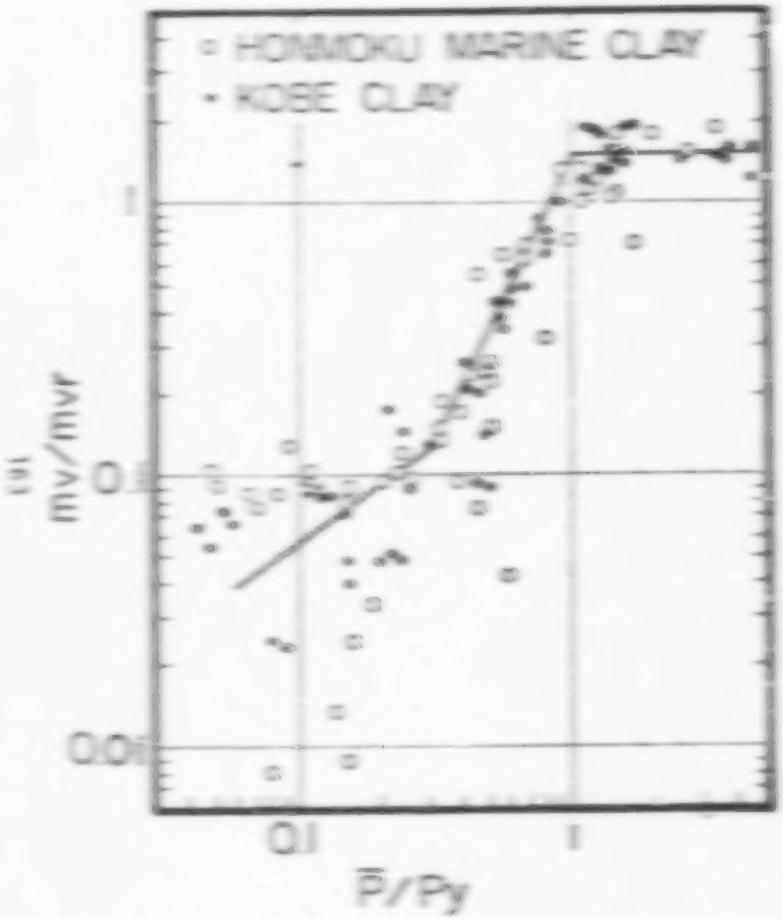


Figure 5. Coefficient of volume compressability of lime treated soils.

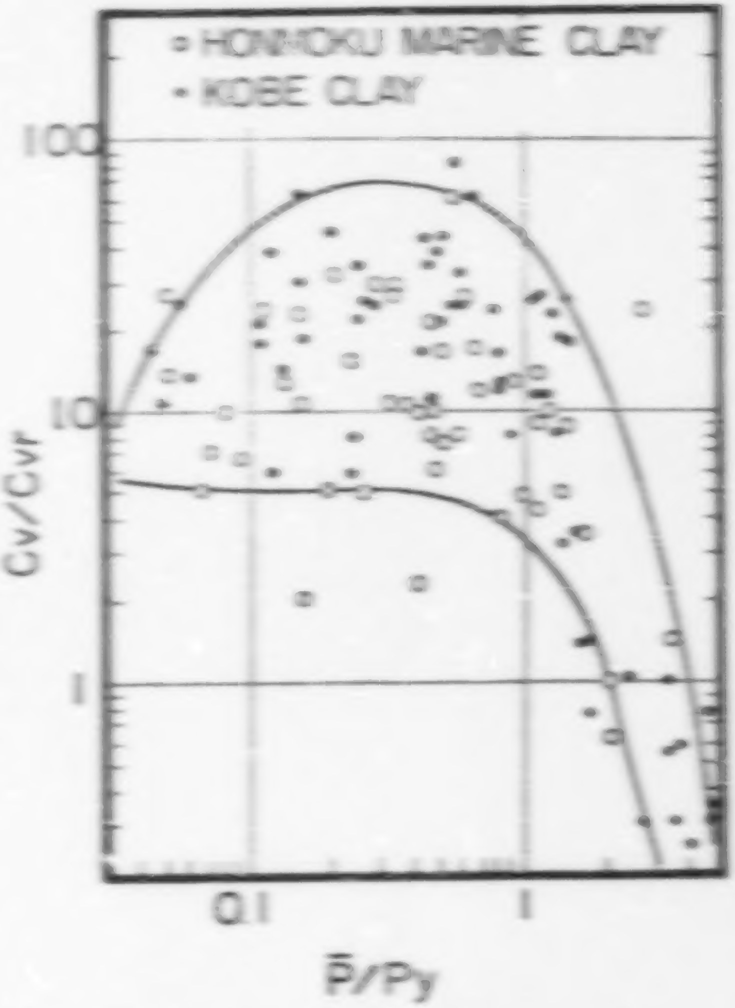


Figure 6. Coefficient of consolidation of lime treated soils.

Test conditions were as follows:

Initial water content, % ··· 100, 200
Agent content in weight relative to the solid weight, % ··· 5, 15
Curing time, days, ··· 3, 7, 21
Curing temperature, ··· 20°C

Test results are compared in Figures 7, 8 and 9 in the form of frequency distributions on a logarithmic scale, where the ordinate is the frequency and the abscissa is the logarithm of the ratio between the two unconfined compressive strengths of soils. These soils are treated under the same conditions with two different stabilizing agents.

Figure 7 compares the stabilizing effect of portland cement and quicklime. There is no significant difference between organic and inorganic soils. On the average, portland cement is more effective by 34%, although the variation is higher in the test cases.

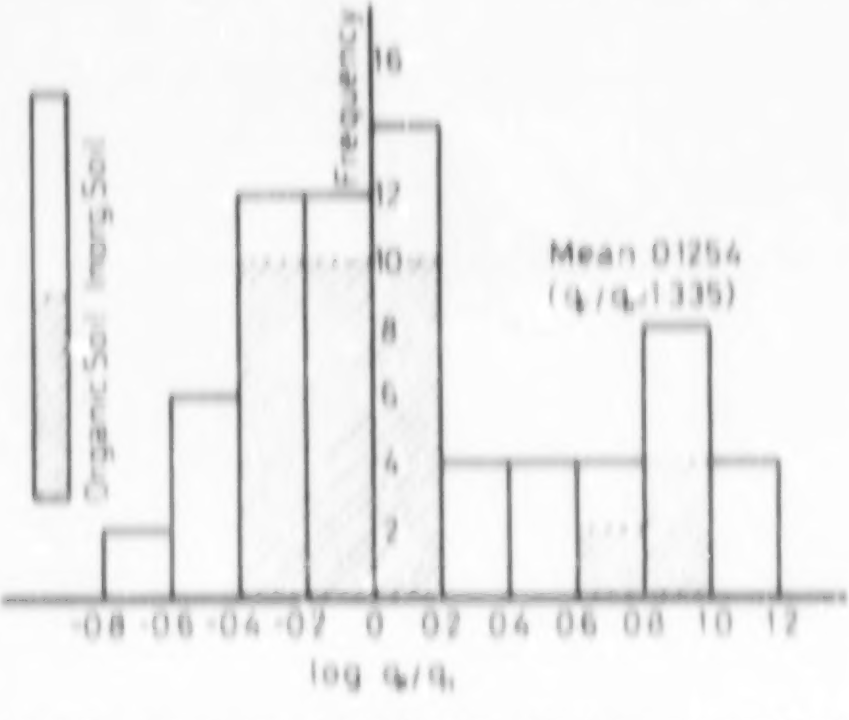


Figure 7. Comparison of stabilizing effect between portland cement and quick lime.

Figure 8 compares the stabilizing effect of portland cement with additive and pure portland cement. In these cases the additive together with portland cement is on the average more effective by 25%.

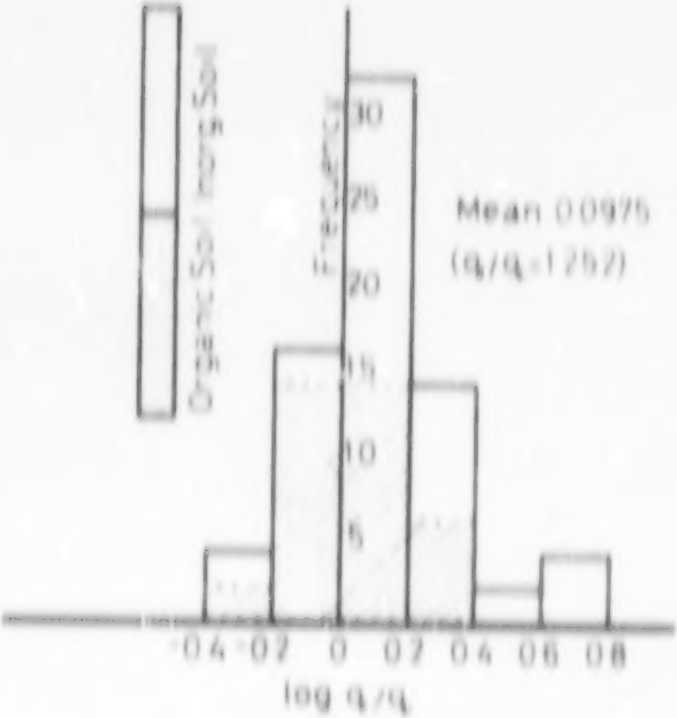


Figure 8. Comparison of stabilizing effect between some cement type agent and portland cement.

Figure 9 shows the stabilizing effect of another additive on organic soil. In these particular cases the additive is less effective by 14%, on the average.

These comparisons show rather wide variation. As previously mentioned, the stabilizing effect of a particular agent depends on both soil and environmental conditions. Figure 10 compares portland cement and quicklime again. In this figure some reliable data other than the above are included. On the average, portland cement is slightly more effective by 12% than quicklime, but with considerable variation among the samples.

Figure 11 compares the stabilizing effect of hydrated lime and quicklime. In this case, it is rather clear that hydrated lime is less effective by about 40%.

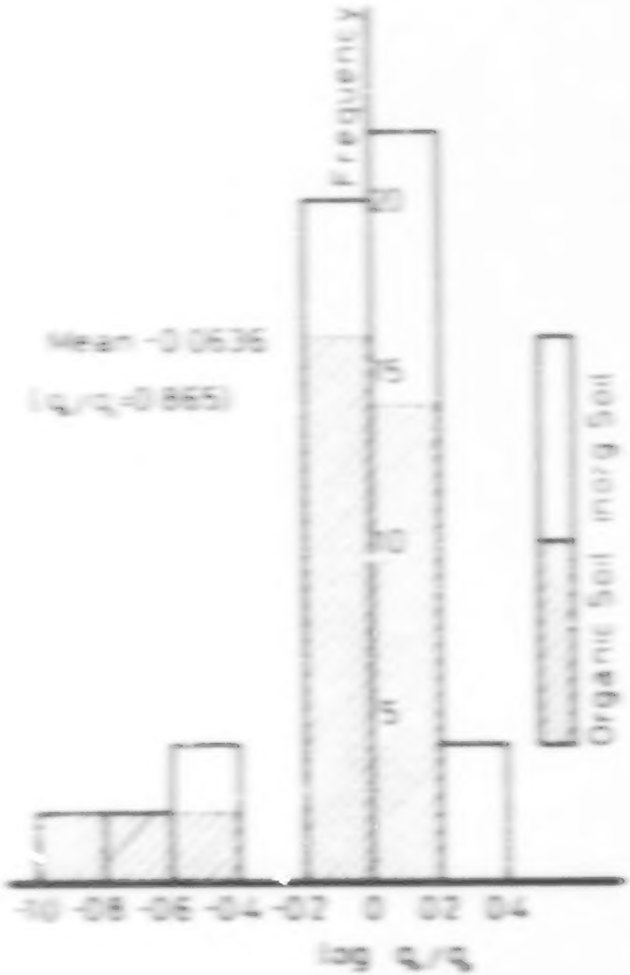


Figure 9. Stabilizing effect of same additive for organic soil.

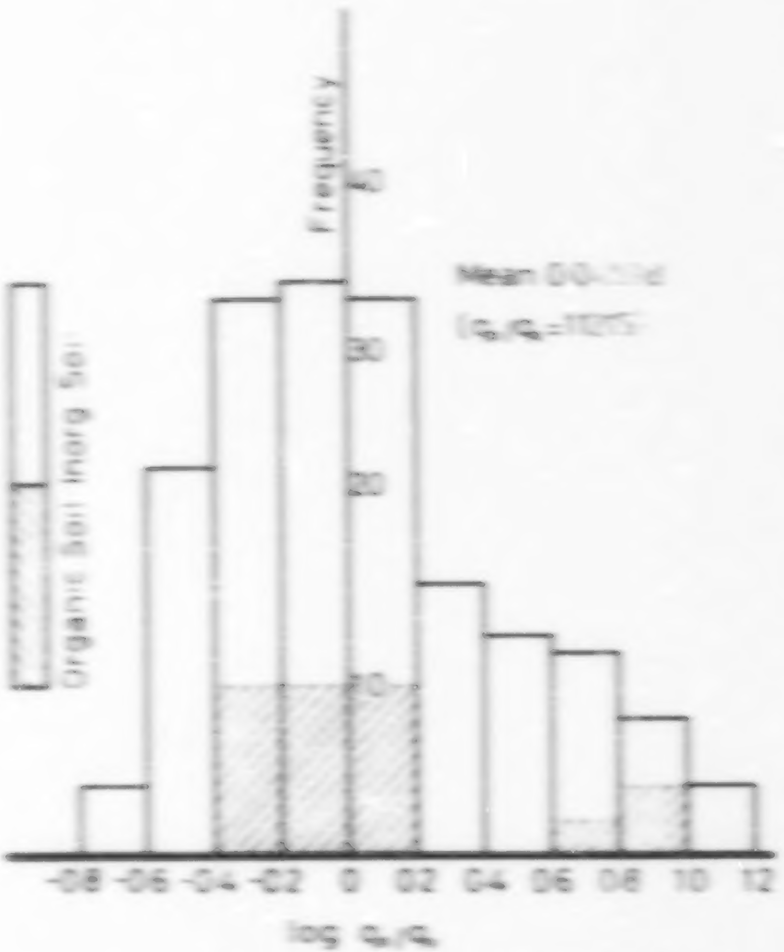


Figure 10. Comparison of stabilizing effect between portland cement and quick lime.

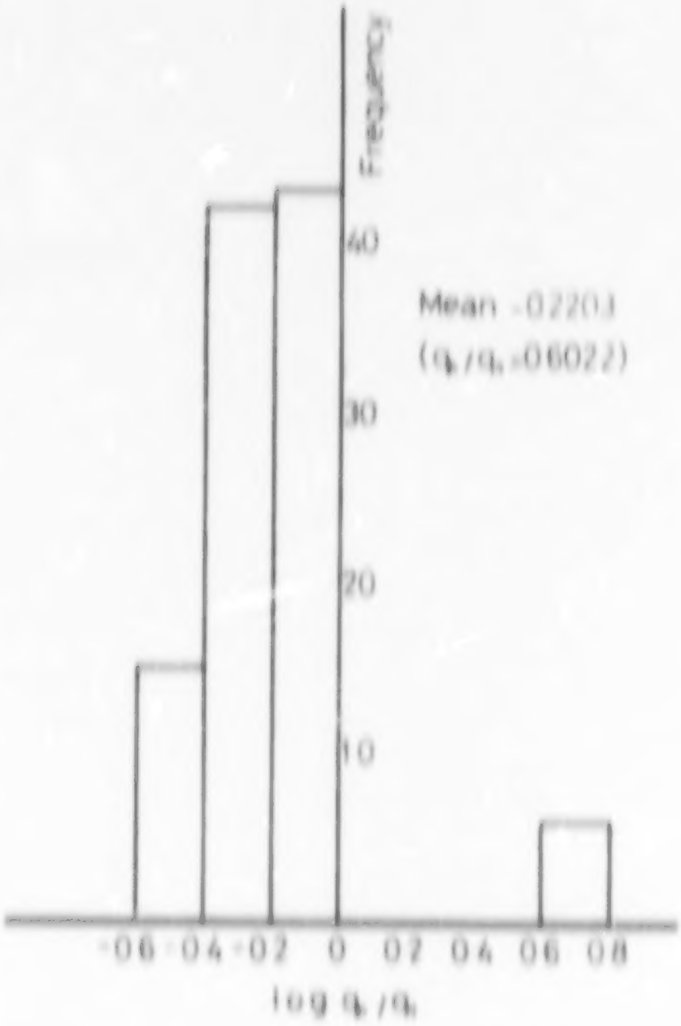


Figure 11. Comparison of stabilizing effect between hydrated lime and quick lime.

CONCLUSION

There are many kinds of chemical stabilizing agents in use. But, their stabilizing effects are subtly influenced by the chemical and mineralogical properties of the soils to be treated and by the physical and environmental conditions of treatment. There exists no single, effective stabilizing agent, i.e., no one agent is suitable for all kinds of soil and all conditions of treatment. In practice every kind of stabilizing agent should be tested for the particular soil and conditions to be treated, and the best agent should be selected for each particular case.

The commercial cost of some special additives makes them impractical, even though they add some degree of increased strength in the stabilization. It is much more reasonable, in practice, to increase the content of the simple stabilizing agents such as quicklime, hydrated lime, portland cement, and slag cement.

REFERENCES

1) Kotani, S. et al. (1974): On the Slag Type Agent for Soil Stabilization, Proc. Japanese Soc. Soil Mech. and Foundation Eng., Vol. 22, No. 5, pp. 75-80 (in Japanese)
2) PHRI, Soil Stabilization Laboratory (1975): Laboratory Testing Manual-- Deep-Lime-Mixing Method for Soil Stabilization ---, 13 p. (in Japanese)
3) Okumura, T., Terashi, M. and Yoshida, T. (1974): Studies on the Engineering Characteristics of Lime Treated Soils (1st Rept.), Proc. 9th Annual Meeting of Japanese Soc. of Soil Mech. and Foundation Eng., No. 225, pp. 893-896 (in Japanese)
4) Okumura, T., Terashi, M. Mitsumoto, T., Yoshida, T. and Watanabe, M. (1974): Deep-Lime-Mixing Method for Soil Stabilization (3rd Rept.), Rept. of PHRI, Vol. 13, No. 2, pp. 3-44 (in Japanese)
5) Okumura, T. and Terashi, M. (1975): Deep-Lime-Mixing Method of Stabilization for Marine Clays, Proc. 5th Asian Regional Conf. on Soil Mech. and Foundation Eng. (Bangalore, India), Vol. 1, pp. 69-75
6) Yanase, S. (1968): Stabilization of Alluvial Clays with Quick Lime (1st Rept.) Rept. of PHRI, Vol. 7, No. 4, pp. 85-132 (in Japanese)

A METHOD FOR DISPOSING OF WASTE WATER AT DREDGED MATERIAL RECLAMATION SITES

E. Satoh*
Japan Dredging and Reclamation Engineering

ABSTRACT

This report introduces a method for disposing of the waste water that continuously flows in huge volumes from dredged materials as they are removed, transported and dumped in reclamation sites. This method is already being put into practice in Japan. It is characterized by its attempt to reduce the quantity of flocculants and to establish a stable and reliable system for disposal of large volumes of waste water by taking advantage of the natural conditions at the site to be reclaimed.

INTRODUCTION

Reclamation using dredged materials requires disposal of the waste water that flows from the reclaimed land. When the dredged materials used for reclamation are polluted, strict controls have to be placed on the toxic compounds; even if the materials are not polluted, it would still be desirable to reduce turbidity to the lowest possible level. The salient point for disposal of waste water derived from reclamation using dredged materials is that the disposal has to be conducted under stable conditions while continuously handling a huge volume of water.

The conventional method of solving this problem focuses on purification of waste water by means of flocculants. This method is quite expensive and also causes concern about the toxic effects of the flocculants. These defects in the conventional method have compelled us to study and find ways of lowering the turbidity to a required level while reducing the quantity of flocculants.

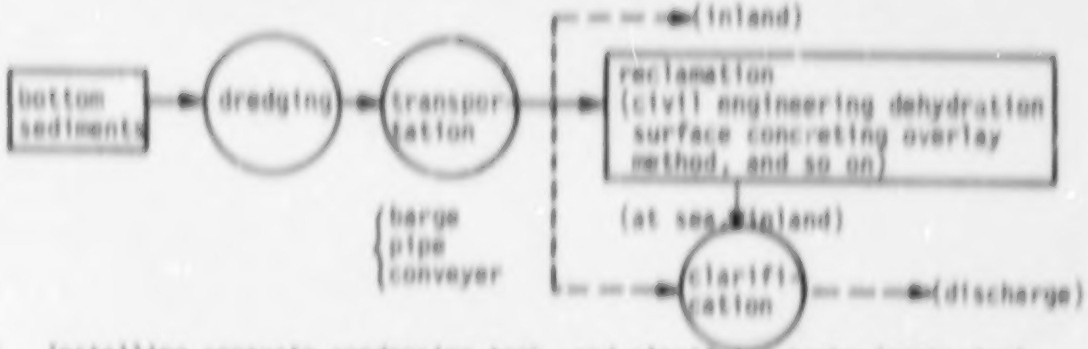
PRESENT STATE OF WASTE WATER TECHNOLOGY

Figure 1 is a flow chart that shows the basic processes of dredging seabed sediments and the use of dredged materials in later reclamation works. These processes are actually in practice in Japan. The materials dredged from the seabed are transported to the reclamation site by a pipeline system, or by other methods. Figure 1-1 represents a method that makes good use of the reclamation area itself as a condensation pond for reclaimed materials, and at the same time, as a clarification pond for waste water. This method

---

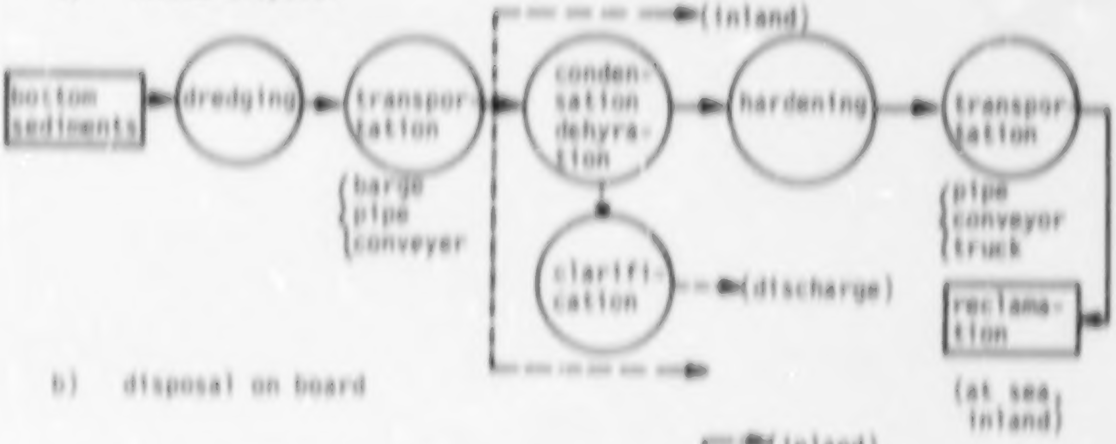
* Toranomon-Kotohirakaikan Bldg. 1 Shiba-Kotohira-cho, Minato-ku, Tokyo 105, Japan

1) Using the reclaimed land as a condensing and clarifying tank


(inland)
bottom sediments
dredging
transportation
reclamation (civil engineering dehydration surface concreting overlay method, and so on)
barge
pipe
conveyer
(at sea, inland)
clarification
(discharge)

2) Installing separate condensing tank and clarifying tanks (mechanical dehydration)

a) inland disposal


(inland)
bottom sediments
dredging
transportation
condensation dehyration
hardening
transportation
barge
pipe
conveyer
clarification
(discharge)
pipe
conveyor
truck
reclamation
(at sea, inland)

b) disposal on board

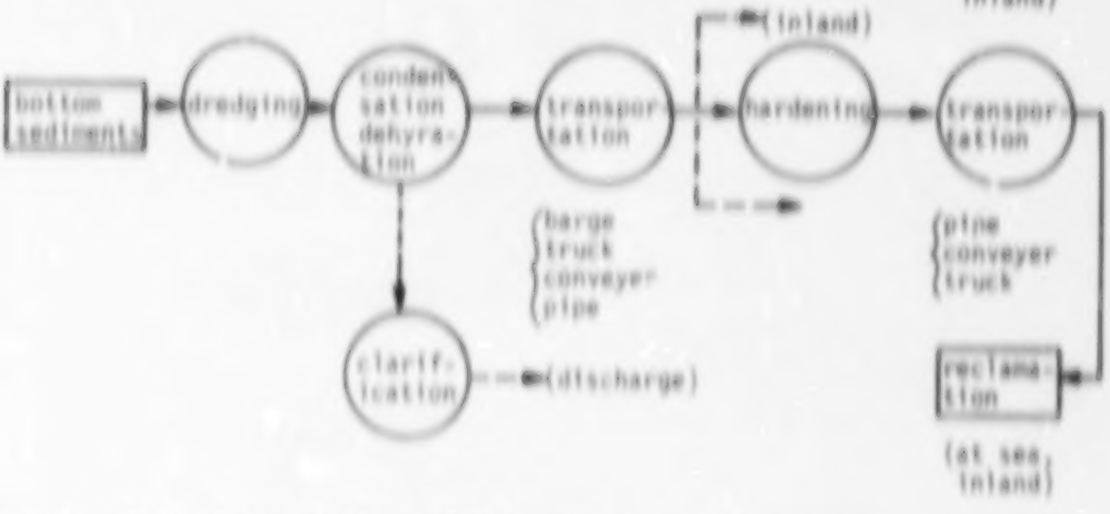

(inland)
bottom sediments
dredging
condensation dehyration
transportation
hardening
transportation
barge
truck
conveyer
pipe
clarification
(discharge)
pipe
conveyer
truck
reclamation
(at sea, inland)

Figure 1. Basic Disposal Flows of Reclamation

is called the "Civil Engineering Dehydration System" because it makes full and effective use of the natural conditions.

By comparison, Figure 1-2 shows the method in which dehydration is done by installing separate condensation and purification tanks. The location where the mechanical dehydration equipment is to be installed must be chosen in each case with consideration of, and in conformity to, the actual conditions of dredging and reclamation. The above mentioned equipment is in some cases installed on shore, but in most cases is ship board. Generally speaking, the method shown in Figure 1-1 is useful for comparatively large scale work, while the one shown in Figure 1-2 is useful for small scale work. The waste water produced by dehydration is duly dreated and then drained away. So far in Japan, the waste water from reclamation of unpolluted dredged materials has been returned directly to the sea without treatment of any kind.

The turbidity of the waste water in question varies in proportion to the scale of reclamation work, the volume of waste water itself, the duration of work, and other conditions. Some records of turbidity obtained through projects completed in the past are shown below (1).

During the initial phase of reclamation	75 - 100 ppm
" middle "	3,000 - 5,000 ppm
" final "	15,000 ppm

Under the present laws and regulations there are no special provisions to restrict the quality of waste water derived from reclamation using dredged materials which are not polluted. However, it is desirable to attain and maintain turbidity at the lowest possible level in consideration of environmental effects on the natural stream water and the sea surrounding the reclamation site. For this purpose, the values cited in Table 1 are largely compiled as a "waste water turbidity standard" and constitute a target to be attained.

Additionally it is quite possible that measures will be undertaken in the future to control waste water disposal for all types of reclamation works.

At present, waste water quality is stringently controlled and restricted in all cases of reclamation using dredged seabed materials which contain toxic heavy metals, organic matter, and oily substances.

The flocculation acceleration method, recently implemented for waste water disposal, consists of simultaneous utilization of both poly-acrylic amide and inorganic flocculants. In order to prevent diffusion of turbid water, physical barriers are in common use.

In cases where waste water quality is highly restricted, rapid filtration equipment is often utilized.

TABLE 1. STANDARDS FOR WASTE WATER DERIVED FROM RECLAMATION

Items	Allowable Bounds
cadmium and its compounds	0.1 mg/l
organic phosphorus compounds	1.0 mg/l
chromium and its compounds	1.0 mg/l
hexa-chromium compounds	0.5 mg/l
arsenic and its compounds	0.5 mg/l
mercury and alkyl mercury and its other compounds	none detected
alkyl mercury	none detected
hydrogen ion concentration (pH)	5.0 - 9.0
biochemical oxygen demand (BOD)	160 mg/l mean 120 mg/l during daytime
chemical oxygen demand (COD)	160 mg/l mean 120 mg/l during daytime
suspended solids (SS)	200 ppm mean 200 ppm during daytime

(notes)

(1) "mean concentration during daytime": average polluted conditions

(2) the Environmental Authority directs that waste water may be discharged at 1/2 to 1/4 of the above levels

Figures 2, 3 and 4 are graphic explanations of the disposal methods for polluted seabed materials. These methods have actually been tried and carried out in Japan. In particular, Figure 2 represents experimental work conducted in the Port of Yokkaichi Municipality and Mie-prefecture, for disposal and removal of bottom soil in waters of the port area. The sediment was polluted with mercury and oily substances. In this experimental work the polluted muddy bottom soil was dredged by suction dredges and then transported by barges to the reclamation site. There, the muddy soil settled leaving the upper water relatively clear. This was disposed of using rapid filtration equipment. This method is like the one shown in Figure 1. A fullscale project for removal and disposal of polluted soil from the Yokkaichi Municipality port is scheduled to begin with a total soil volume of 2,200,000 m^3 to be dredged in the latter half of 1976. Completion will take approximately 2 years.

Figure 3 shows the dredging work at Lake Biwa, Shiga prefecture, which involves disposal of pulp sludge containing PCB. This is an example of a relatively small scale project. In this case, dredged sludge was dehydrated by a filter type dehydrator installed in the on-site plant. The dehydrated sludge was solidified by means of cement while the waste water was purified by flocculants and then drained away. This method corresponds to Figure 1-2a.

Figure 4 shows a dredging project now in progress in the Seto Inland Sea which is handling seabed soils polluted by organic matter. This work is being undertaken as a measure to minimize the effects of the red tide. The muddy polluted soil, after being dredged, is brought to the disposal vessel which is moored in the vicinity of the dredging area. Thickening tanks, dehydrators and quick filtration equipment are all installed aboard the disposal vessel. The polluted soil, once dehydrated, is transported ashore and then solidified by cement. The filtered waste water is drained into the sea. This method is shown in Figure 1-2b.

The standards of waste water quality set by regulation are shown below.

Fig. 2:	Yokkaichi Harbor	
	SS	30 ppm
	Normal-hexane soluble substances (daily average)	2 ppm
Fig. 3:	Lake Biwa	
	SS	40 ppm
Fig. 4:	Seto Inland Sea	
	SS	10 ppm

CIVIL ENGINEERING MANAGEMENT OF WASTE WATER

Until recently in Japan, dredging and reclamation of land required that the reclamation site be enclosed by a simple wooden stockade from which all the waste water from the spoils drained through spillways. Muddy soil and water which spilled from the wooden stockade caused turbid water to spread.

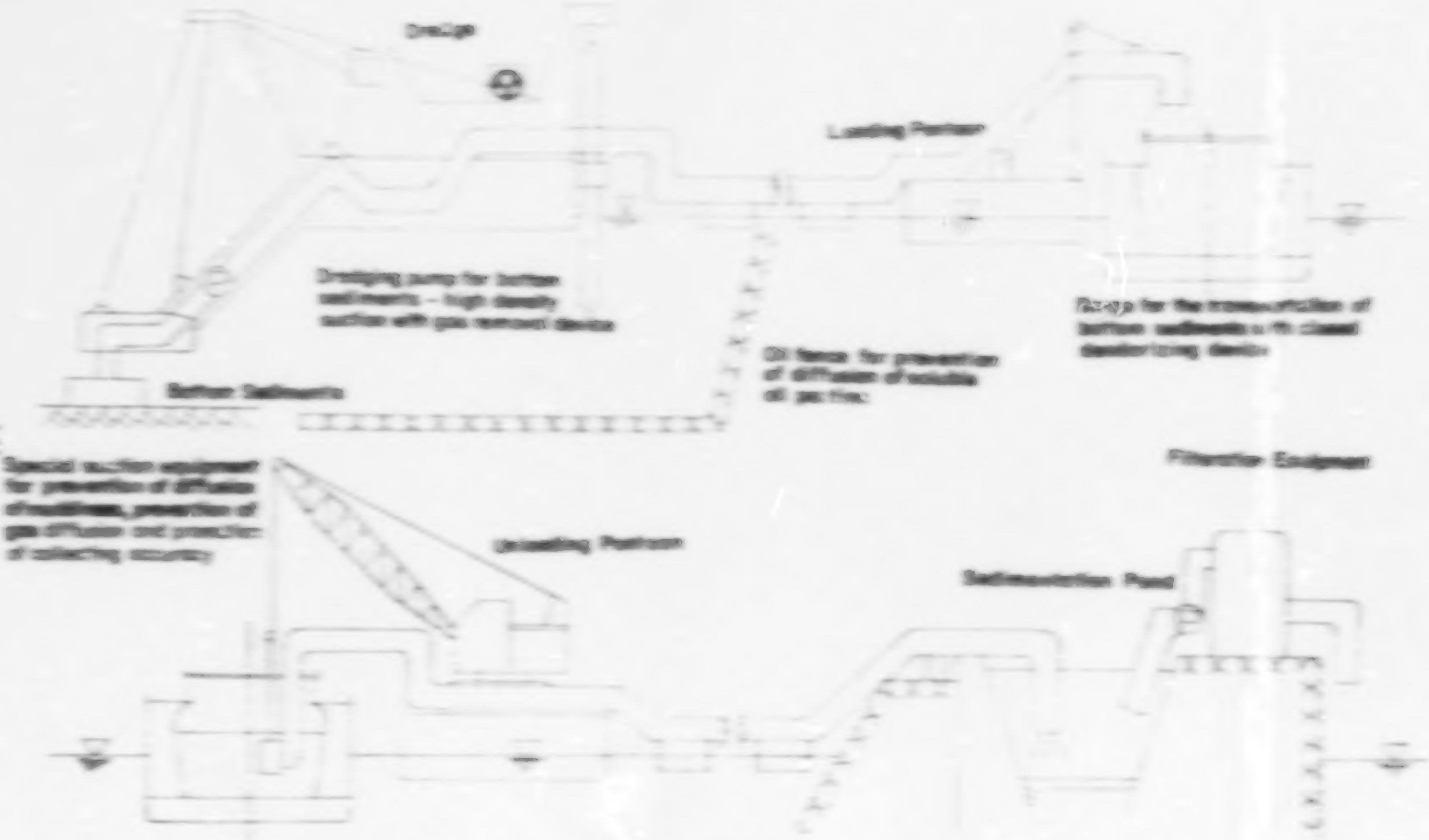
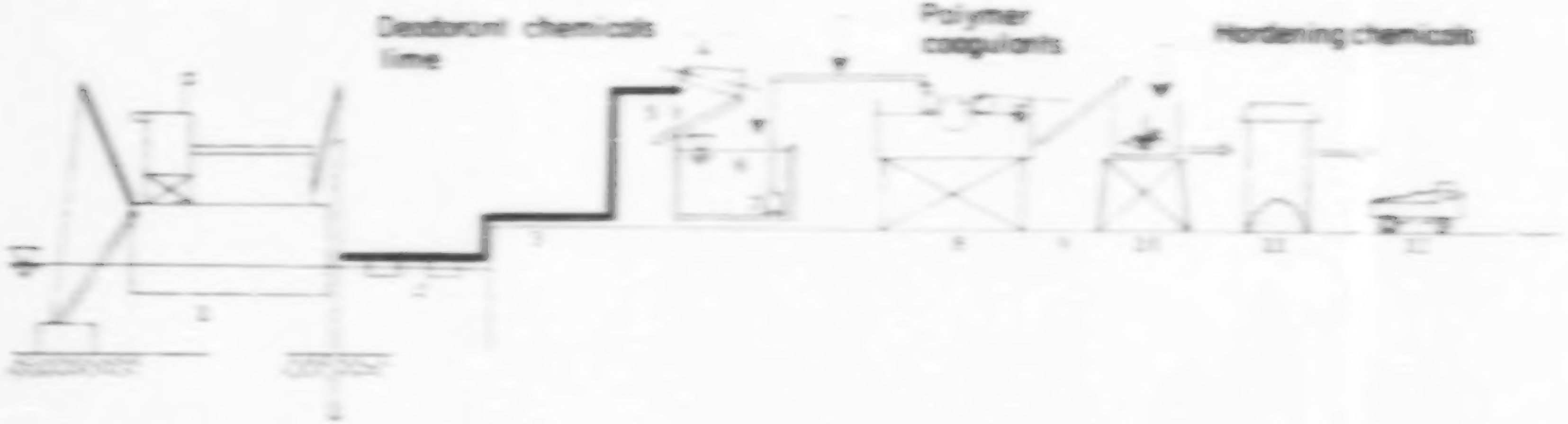
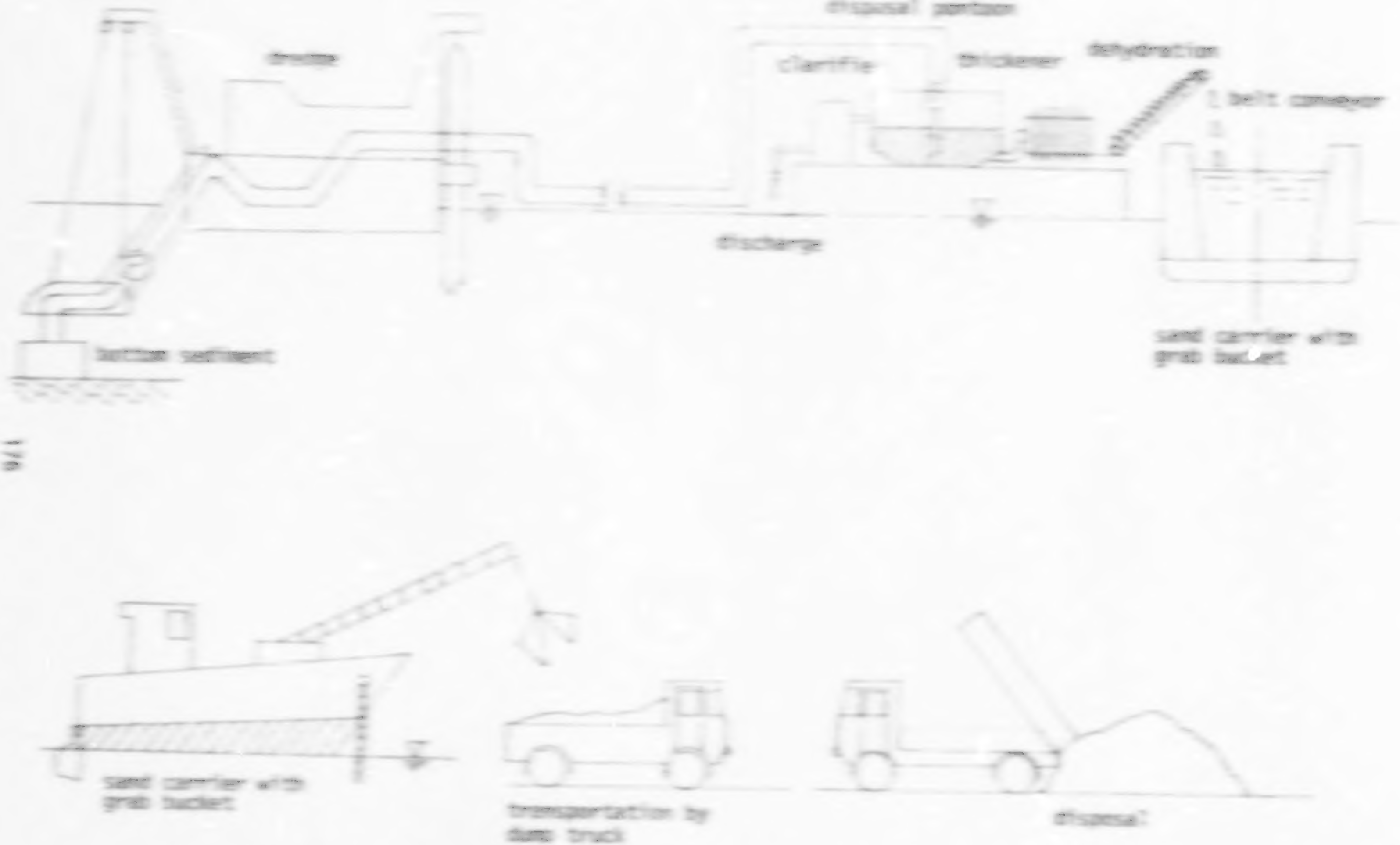

Dredge
Loading Pontoon
Dredging pump for bottom sediments – high density suction with gas removal device
Oil fence for prevention of diffusion of volatile oil particles
Barge for the transportation of bottom sediments with closed deodorizing device
Bottom Sediments
Special suction equipment for prevention of diffusion of turbidness, prevention of gas diffusion and promotion of collecting accuracy
Unloading Pontoon
Sedimentation Pond
Filtration Equipment

Figure 2. An example of reclamation using "Civil Engineering Dehydration."


Deodorant chemicals
lime
Polymer
coagulants
Hardening chemicals

1. Dredge for bottom sediments
2. Floating line
3. Inland pipe line
4. Rotary screen
5. Belt conveyer
6. Cushion tank
7. Submergible sand pump
8. Proportional filter
9. Belt conveyer
10. Mixer
11. Warehouse
12. Dump truck

Figure 1. An example of reclamation using mechanical dehydration with inland disposal.



dredge
disposal pontoon
clarifie
thickener
dehydration
belt conveyor
discharge
bottom sediment
sand carrier with
grab bucket
sand carrier with
grab bucket
transportation by
dump truck
disposal

Figure 4. Another example of reclamation using mechanical dehydration and inland disposal.

Because of the large volumes of silt and clay which were released, the local inhabitants and the general public complained about this method of reclamation.

Therefore, in compliance with public opinion, and prior to the commencement of a project, bulkheads are usually erected to enclose most of the reclamation sites. With this technique almost the entire volume of waste water brought into the area is spilled outside of the bulkheads through spillways and consequently drains to the adjacent water. Therefore, the quality of the waste water flowing through the spillways has become the focus of a water quality problem.

Japanese reclamation projects which make use of quality non-polluted material are done with suction dredges equipped with dredging pumps of around 4,000 ps. This means that, including rainfall, waste water would flow through the spillways at an average rate of 6,000-8,000 m^3 per hour. Dredging of polluted muddy sediments that contain toxic substances require waste water disposal at the rate of 1,000-2,000 m^3 per hour. The point is that the waste water arising from reclamation using dredged materials is a huge volume that has to be dealt with constantly.

In using the bulkhead method it is easy to provide a large settling pond inside the enclosure in the initial phase of the work. However, the pond would be reduced in capacity as the work progressed and would lead to increased waste water turbidity over the spillways. Rainfall would also influence the volume of the waste water at the same time. Thus, the fluctuation in both waste water turbidity and volume constitutes an additional characteristic of the "Civil Engineering Dehydration Method."

Grading of the soil to be dredged is also related to waste water quality. The greater the proportion of minute silt and clay in the dredged material the more difficult the disposal work will be.

In actuality, this waste water disposal method has been applied to reclamation work using normal quality soils and used only in cases where the soils were of soft quality and mostly composed of clay and silt. In these cases the waste water contained a substantial volume of clay.

There are two particular methods which have been applied in these cases. The first is to inject poly-acrylic amide inside the pipe that conveys the dredged soil to the reclamation area with subsequent flocculation and separation of soil grades occurring over the entire spoil area. The second method is the same as the preceding method up to the process of conveying the dredged soil. Once the dredged soil arrives in the reclamation area it is placed in a smaller enclosure within the larger reclamation area. This is where it is concentrated by flocculation and separation processes.

The waste water disposal method which depends wholly upon poly-acrylic amide is open to question. The doubt arises in the application of the flocculants. The method as explained above requires instantaneous flocculation and separation. The direct application of the chemicals to highly turbid water allows insufficient mixing between the chemicals and the muddy water unless huge doses of poly-acrylic amide are used. Some actual cases report an average use ranging from 10 to several hundred grams of poly-acrylic amide per 1000 kg of reclaimed soil (dry weight basis). Therefore, these present methods not only subject the area to the toxic effects of poly-acrylic amide, but also are not economical.

In cases where the waste water density and its volume fluctuate, the dose of chemical is figured by assuming the worst possible conditions. This sometimes compels an unnecessary quantity of chemicals to be used needlessly. In cases where polluted muddy seabed soil is handled, stringent water quality standards often require an excessive application of flocculants.

Thus, while the original motivation for experimenting with new methods for waste water disposal in reclamation is undoubtedly an attempt to minimize turbidity it is, at the same time, aimed at reducing the consumption of poly-acrylic amide. With this point of view in mind a group of engineers, under the leadership of the author, have conducted the following studies and investigations.

Development of a Method of Settling and Consolidation of Dredged Materials in an Enclosed Reclamation Area Without Using Flocculants

This method consists of two steps. The first step is to utilize effectively the water-bearing area of the reclamation site for a sedimentation pond. There is a limited water volume in the reclaimed area, especially in the final stages of reclamation. There are also many complex currents in the sedimentation pond, e.g. surface currents, short cut flows and wind waves. In addition, the settling time of minute particles is considerable. Because of these problems the capacity of a pond for sedimentation and consolidation of the dredged material is limited. This makes it very difficult to estimate the capacity of a sedimentation pond. In spite of these difficulties, we consider it worthwhile to try to utilize the full capacity of any sedimentation pond. In order to utilize the reclaimed area for primary sedimentation it is important to consider plants growing in the reclaimed area (such as reeds), the plan for reclamation procedures, and the plan of discharge pipelines. This first step is only to decrease the turbidity of waste water.

The second step of this study is intended to accelerate the settling and precipitation of the particles in the waste water. For this reason, an attempt was made to take advantage of the location where the waste water was to be discharged from the reclaimed area as a second sedimentation pond. This water collecting sedimentation pond is based on our idea to settle and precipitate minute particles without using flocculants.

This sedimentation pond, which we call the "Trough Spillway," is discussed later in this report along with an explanation of its structural and test results. This sedimentation pond is aimed at promoting settling and precipitation without using electric power or mechanical devices, and is also aimed at making good use of natural conditions. Because of this we call this technique the "Waste Water Disposal Applied Civil Engineering Method" to distinguish it from the conventional "Civil Engineering Dehydration Method."

A Method of Flocculation and Separation of Mass-Waste Water Using Temporary Rectifying Sedimentation Ponds in Reclaimed Areas in Combination with the Waste Water Controlling System

When reclamation work uses dredged bottom sediments containing noxious materials, there are severe restrictions on waste water turbidity and quality. We often cannot attain the standard using only natural sedimentation and separation. In this case the use of flocculants is necessary. The temporary rectifying sedimentation pond, located in the reclaimed area in combination with the controlling system, is a rational system to continuously dispose of a huge volume of waste water.

The waste water flowing though the reclaimed area is introduced into a sedimentaion pond of required volume. There chemicals are added to the water and its currents are "rectified" by means of baffle boards in the pond. Actual instances where this system was applied are discussed in the following section. The rectifying sedimentation pond system is not effective without being combined with a waste water control system as discussed above. Properly done, a stable and economical mass waste water disposal system is realized by adjusting the amount or chemicals in proportion to the water quality and quantity flowing into the rectifying sedimentation pond. By controlling the discharge rate and chemical dose, water quality is precisely adjusted to meet the standards.

AN EXAMPLE OF THE RECTIFYING SEDIMENTATION POND METHOD

Bottom sediments containing cadmium in Shimonoseki City, Yamaguchi Prefecture, were dredged from 1973 to 1974. Regulation of the waste water standards are as follows:

SS	less than 20 ppm
Cadmium	less than 0.005 mg/l
pH	5.8 to 8.6

Structure

Figures 5 and 6 show the general plan of the work site and a side view of the rectifying sedimentation pond. Dredged slurry is transported via pipeline to the reclamation area, which is enclosed by a watertight structure.

The disposal point of the dredged slurry is the place farthest from the reclamation area behind the fence (see Figure 5). Waste water is conducted into the rectifying sedimentation pond. Poly-acrylic amide is added and the pH value is adjusted using chemicals, such as hydrochloric acid, which are added to the waste water at the entrance of the sedimentation pond. Steel "rectifying plates" are baffles set into the sedimentation pond to regulate currents in the waste water. The seepage ratio of the rectifying plate is 3.1%. The quantity of waste water is 270 m^3/hr, the mean velocity of the rectifying current is 3 mm/sec, and the residence time is around 10 hours. The volume of the flocculants used is 0.9 g/m^3 and 7 cc of 30% hydrochloric acid is used as the neutralization chemical.

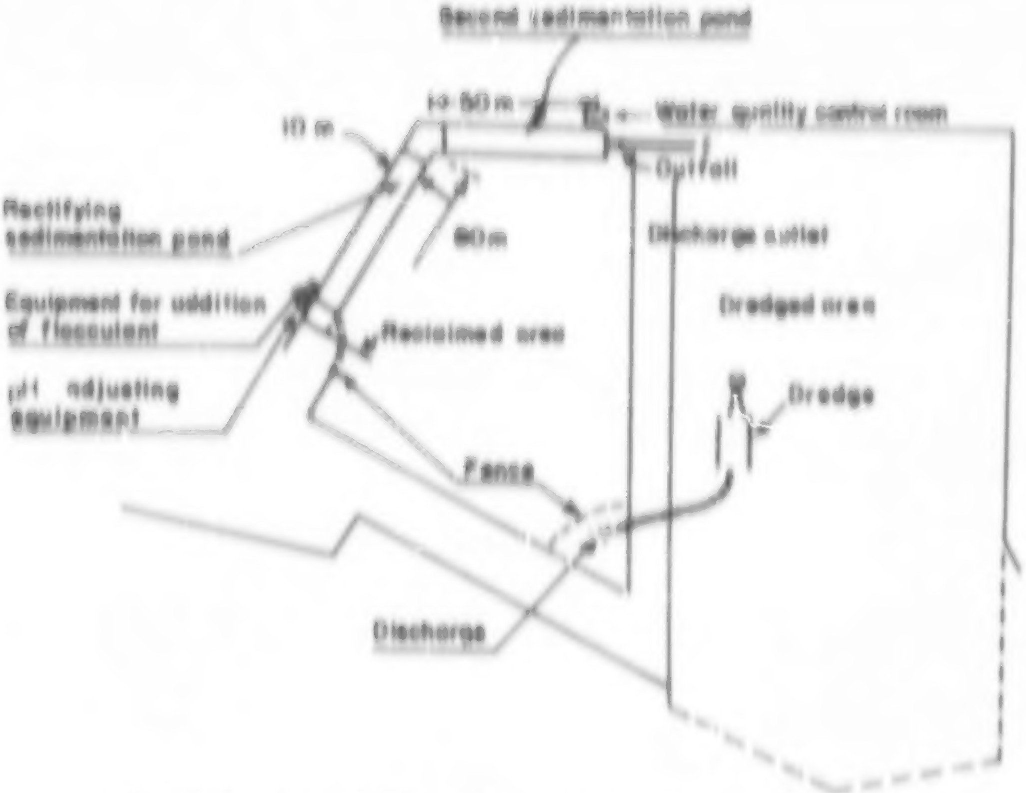

Second sedimentation pond
← 50 m →
Water quality control room
10 m
Outfall
Rectifying sedimentation pond
80 m
Discharge outlet
Equipment for addition of flocculent
Dredged area
Reclaimed area
pH adjusting equipment
Dredge
Fence
Discharge

Figure 5. Rectifying sedimentation pond as used with dredged materials disposal for land reclamation.

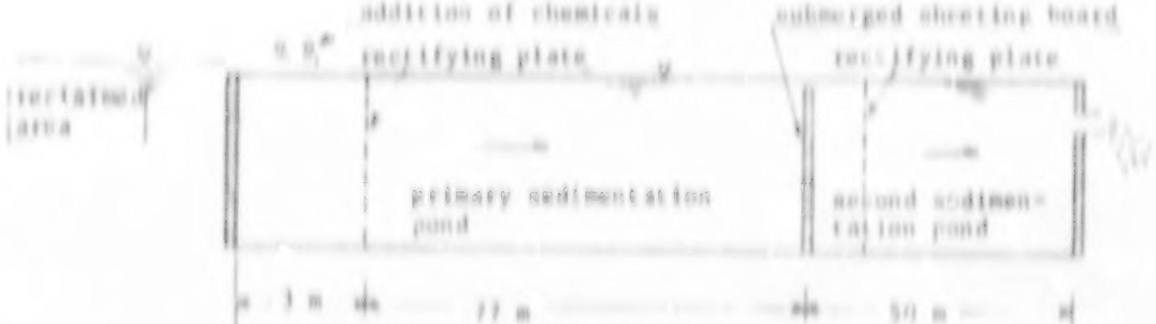

addition of chemicals
submerged sheeting board
0.5 m
rectifying plate
rectifying plate
reclaimed area
primary sedimentation pond
second sedimentation pond
3 m
77 m
50 m
130 m

Figure 6. Side view of rectifying sedimentation pond.

The sedimentation pond is a two-stage structure; the partition wall, or wier, is of the "submerged sheeting board" type. Figure 7 shows the control system for the waste water and Figure 8 the flow chart the for the pH adjusting system.

Test Results

In order to examine the effectiveness of the rectifying sedimentation pond, SS values of the waste water were tested constantly at the pond entrance, and at 3 meters and 80 meters from the entrance of the pond. Table 2 shows an example of the test results.

Evaluation

As shown in Table 2, the average SS value at the sedimentation pond entrance is 26.5 ppm, while the average SS value of the waste water passing through the sedimentation pond is 9.3 ppm. Generally, when the SS value is small, the components of suspended solids almost always consist of minute particles which are difficult to settle and precipitate. For the rectifying sedimentation pond, however, the removal ratio of 65% achieves the SS values desired.

TABLE 2. SUSPENDED SOLIDS IN THE RECTIFYING SEDIMENTATION POND

Measuring Point	Measurement (ppm) Place	Average during 70 days (pneuma pump) Turbidity (ppm)	SS	Removal Ratio (%)	Average during 11 days (centrifugal pump) Turbidity (ppm)	SS	Removal Ratio (%)
1	at flowing entrance	25.2	14.1	---	49.6	26.5	---
2	3m downstream from flowing entrance	18.5	10.7	26.6	31.7	17.4	36.1
3	80m downstream from flowing entrance	16.1	8.9	13.6	20.7	11.8	22.3
4	130m downstream from flowing entrance	12.9	7.6	8.7	15.8	9.3	9.9
				48.8			68.8

Figure 9 shows that in the rectifying sedimentation pond almost all the soil is accumulated in the first area. This is the whole point of the two-stage sedimentation pond.

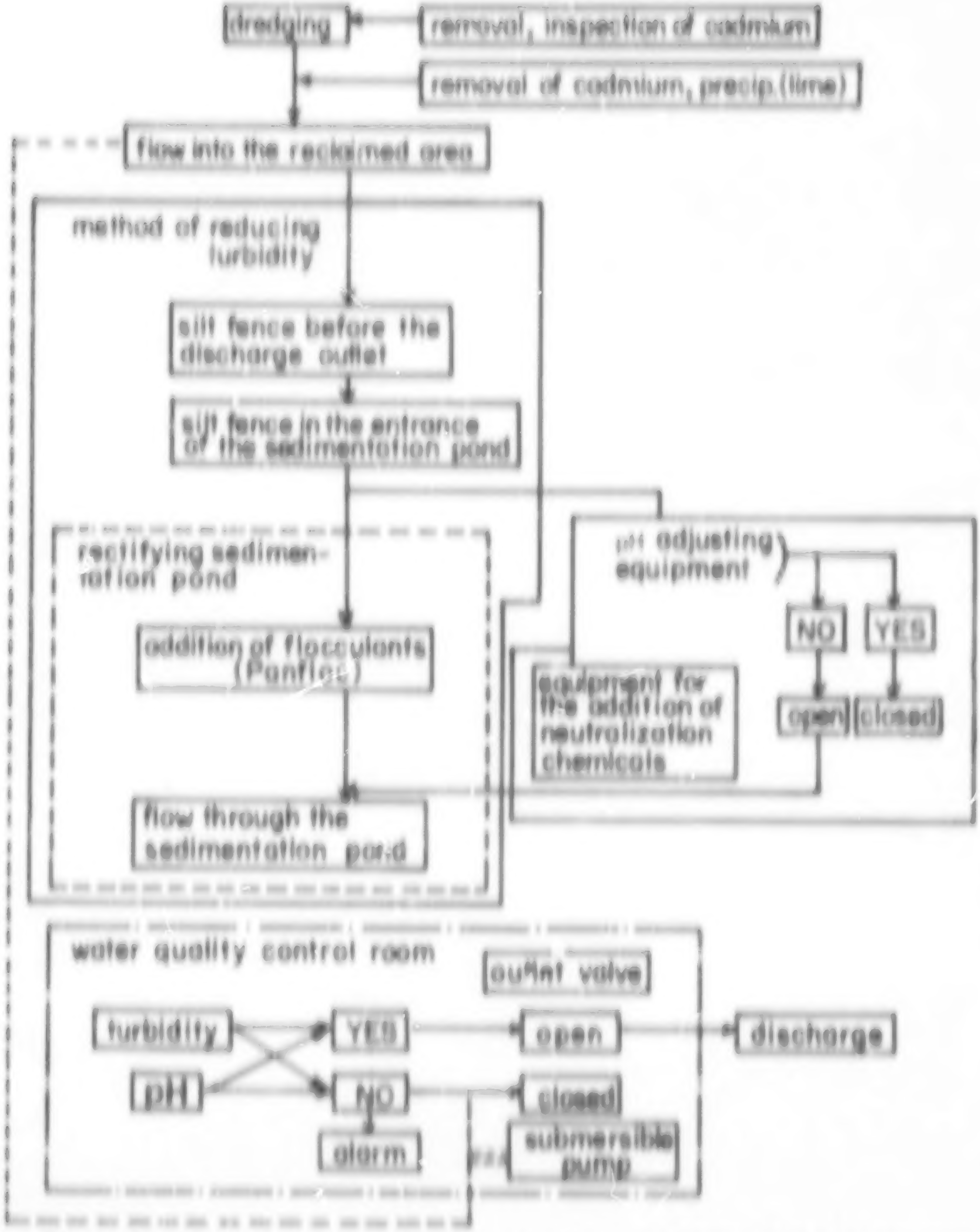

dredging
removal, inspection of cadmium
removal of cadmium, precip.(lime)
flow into the reclaimed area
method of reducing turbidity
silt fence before the discharge outlet
silt fence in the entrance of the sedimentation pond
rectifying sedimentation pond
addition of flocculants (Panfloc)
flow through the sedimentation pond
pH adjusting equipment
NO
YES
open
closed
equipment for the addition of neutralization chemicals
water quality control room
outlet valve
turbidity
pH
YES
NO
alarm
open
closed
submersible pump
discharge

Figure 7. Waste water controlling system in rectifying sedimentation pond.

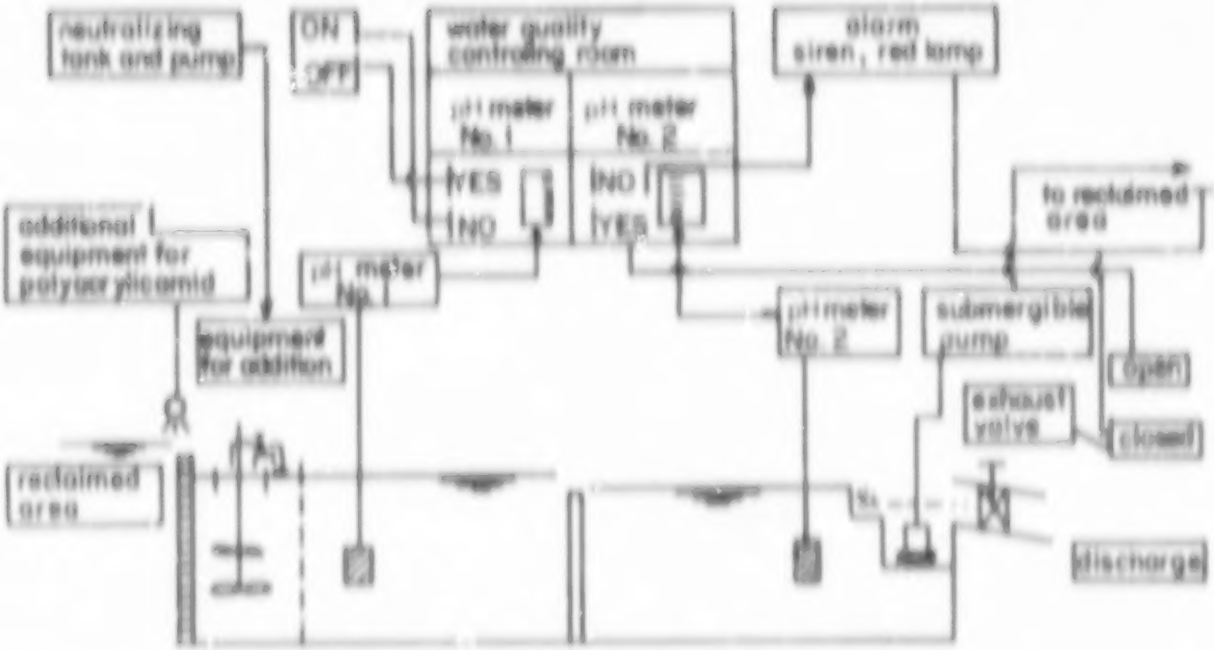

neutralizing tank and pump
ON
OFF
water quality controlling room
pH meter No.1
pH meter No.2
YES
NO
NO
YES
alarm siren, red lamp
to reclaimed area
additional equipment for polyacrylicamid
pH meter No.1
equipment for addition
pH meter No.2
submergible pump
open
exhaust valve
closed
reclaimed area
discharge

Figure 8. pH adjustment flow in rectifying sedimentation pond.

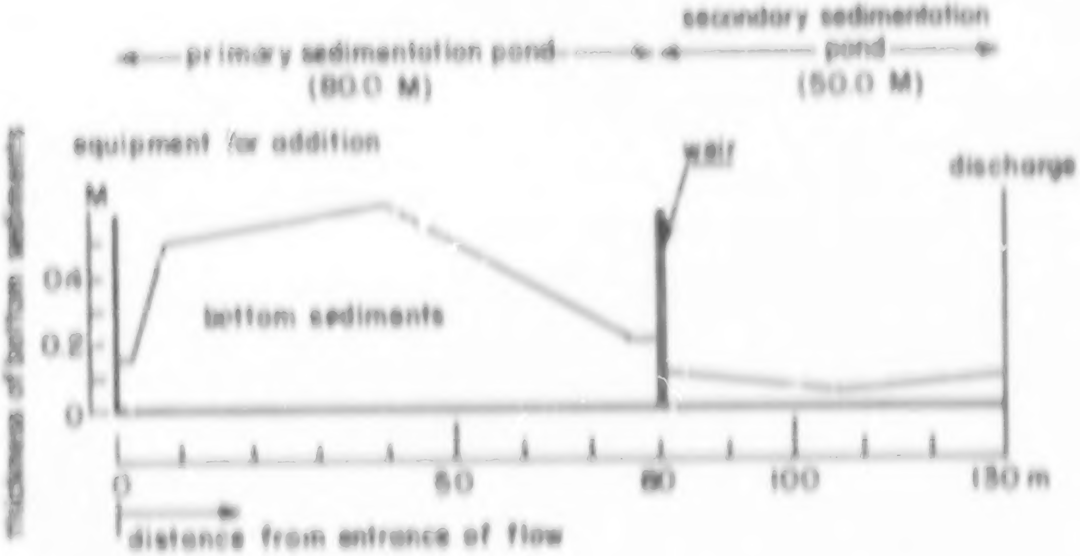


Figure 9. Soil accumulation conditions in rectifying sedimentation pond.

EXAMPLE OF THE TROUGH SPILLWAY METHOD IN USE

The trough spillway method was developed through fundamental experiments. It has been applied in the reclamation projects in Kumamoto Prefecture in 1975 and 1976. Here, the materials to be reclaimed were not polluted. However, the sediment contained large amounts of silty and clayey materials which would cause turbidity. This is why the spillway method was applied at that time. The quantity of water to be disposed of was 5000 m^3/hr. No flocculants were applied in this work.

Structure of Trough Spillway Method

Figure 10 shows the structure of the trough spillway. This is built at the outlet portion of the waste water sedimentation pond in the reclamation area. The spillway consists of the following sections:

Submerged sheeting boards	: 2
Curtain walls inside boards	: 3
Horizontal filters inside walls	: 4
Trough floating on the water	: 1

A 10 cm thick flow of waste water passes over the sheeting board which acts as a wier. This overflow collects the surface water. Then, in the second stage, the waste water which overflows into the inside of the boards is changed to a vertical ascent current within the curtain walls. The average ascending velocity of this vertical current is 3 mm/sec, which is equivalent to the natural settling velocity of 55 μm diameter soil particles. In the third stage the ascending waste water passes through the horizontal filters which have an effect of stopping the upwelling of soil particles. The apparent density difference can be seen on both sides of the horizontal filters. In the last stage, the waste water is discharged to open water outside of the reclamation area via a 1,000 m long trough with an overflow depth of about 6 mm. To maintain efficient sedimentation the materials which accumulated on the bottom of the trough spillway were transported to the reclamation area with a submersible pump. Figure 11 shows the installation of the trough spillway structure.

Test Results of the Trough Spillway Method

To evaluate the trough spillway method turbidity values for the waste water were sampled in the overflow of the submerged sheeting board and the discharged water from the trough spillway. These are shown in Table 3. On the whole, turbidity removal efficiency is high for the waste water flowing into the trough spillway system. Figure 12 shows some of the records obtained by the continuous turbidity recorder. Fluctuations of turbidity values for the inlet waste water are large, while fluctuations in turbidity for the waste water outlet from the trough spillway are small. Figure 13 shows the particle size distributions for mud sampled at several points. This shows that the percentage of minute particles increases from the first to the last stages.

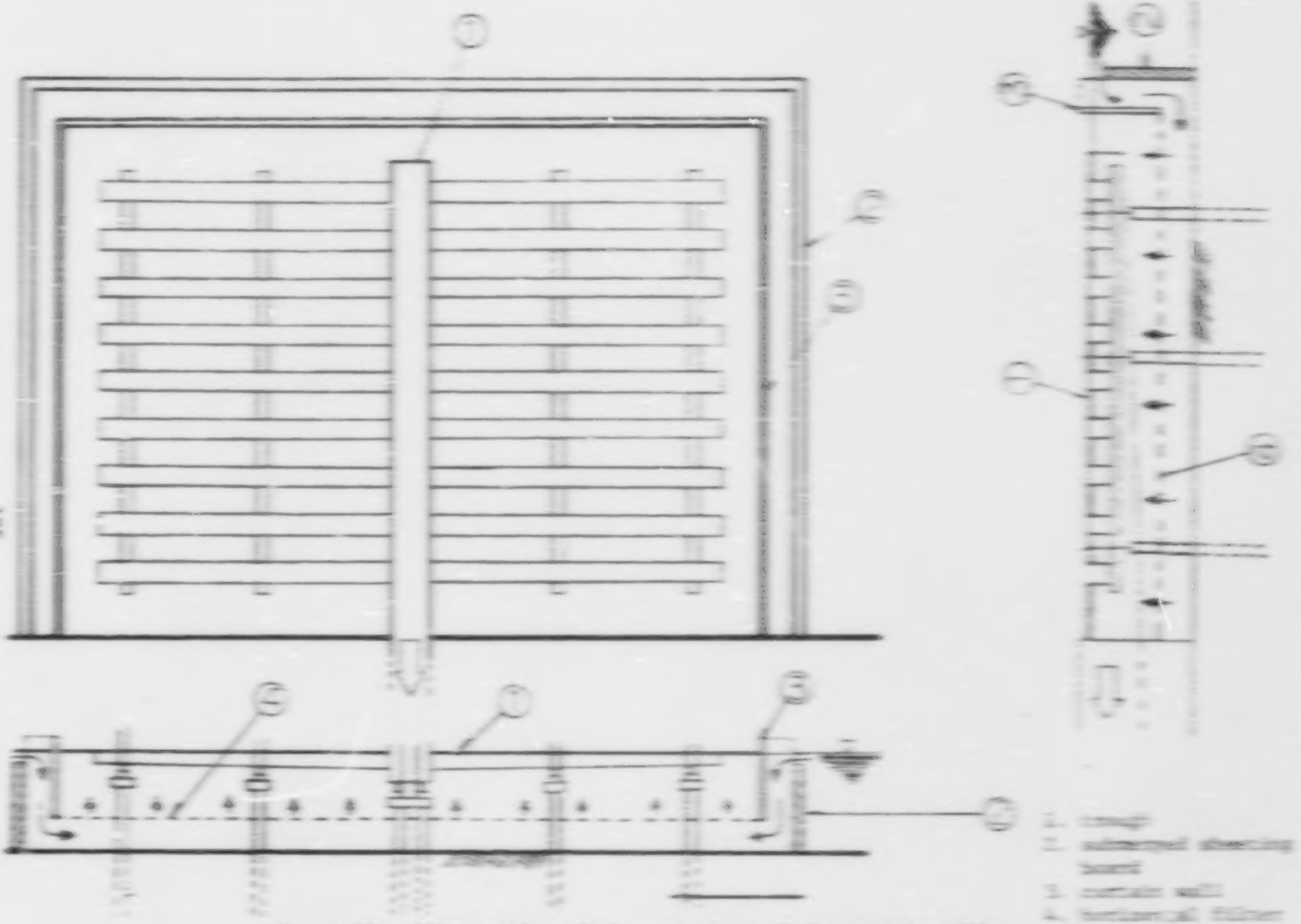

①
②
③
④
1. trough
2. submerged sheeting board
3. curtain wall
4. horizontal filter

Figure 18. Illustration of the structure of the trough spillway.

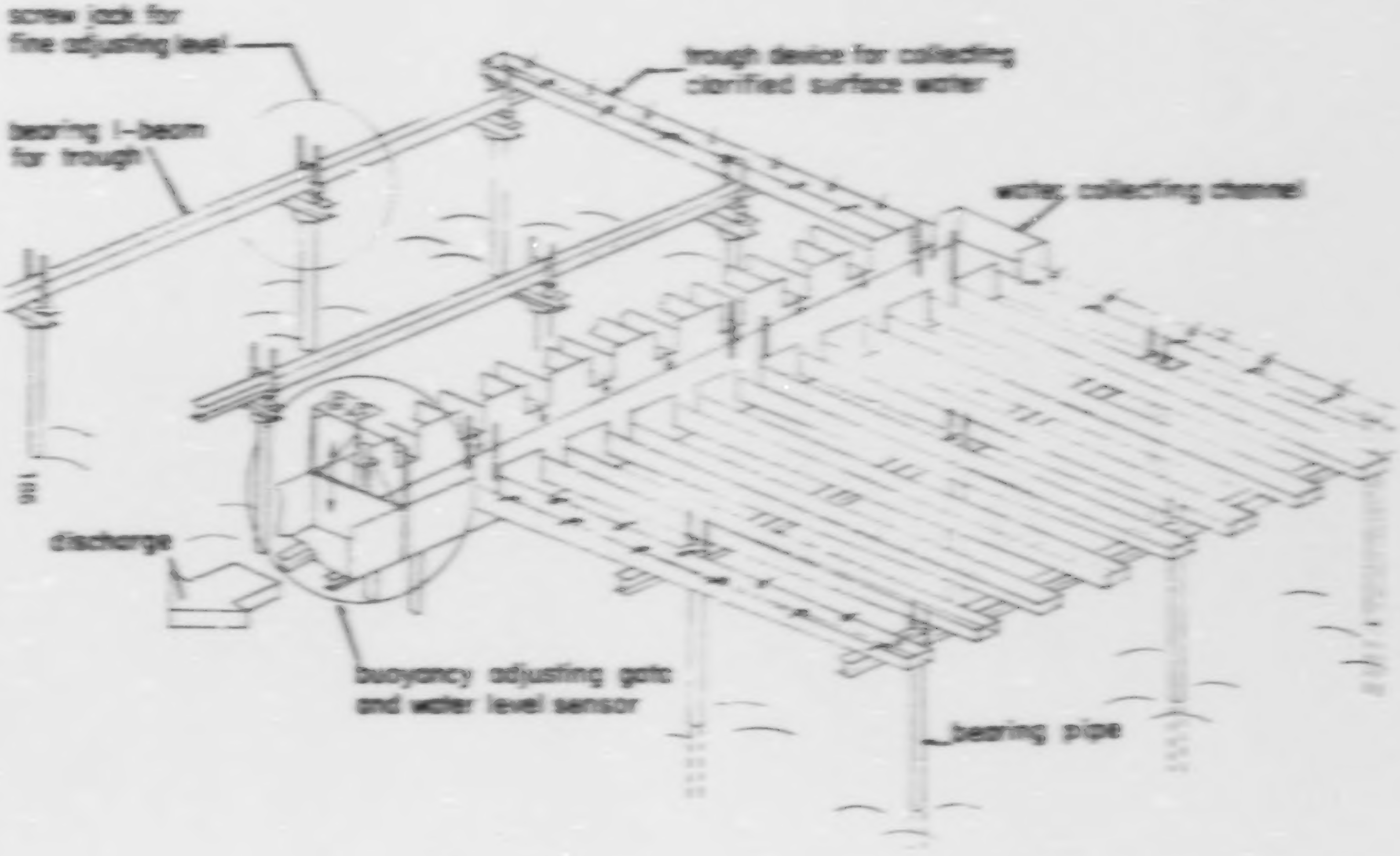

screw jack for
fine adjusting level
trough device for collecting
clarified surface water
bearing I-beam
for trough
water collecting channel
discharge
buoyancy adjusting gate
and water level sensor
bearing pipe

Figure 11. Structure with set troughs.

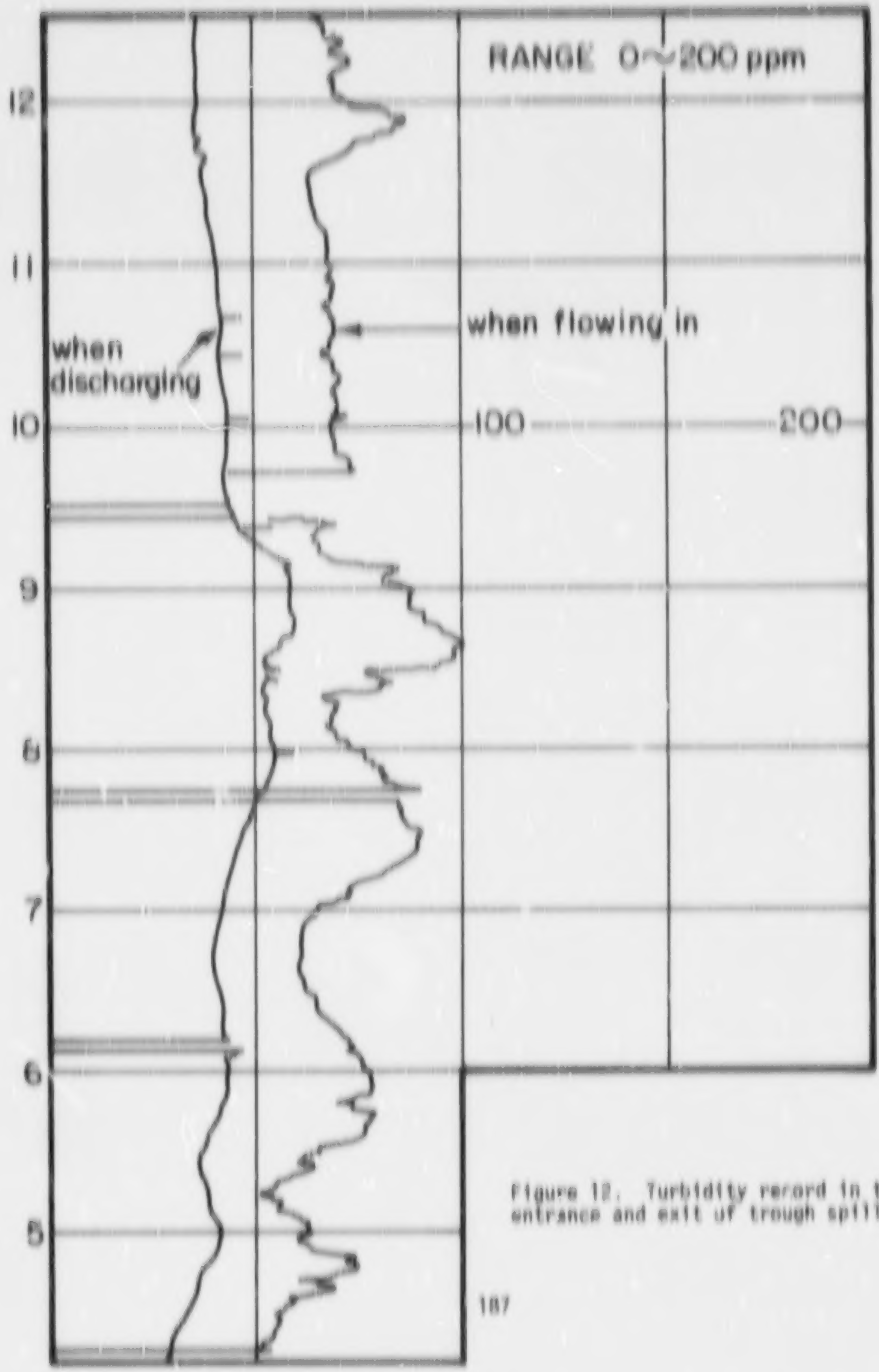


Figure 12. Turbidity record in the entrance and exit of trough spillway.

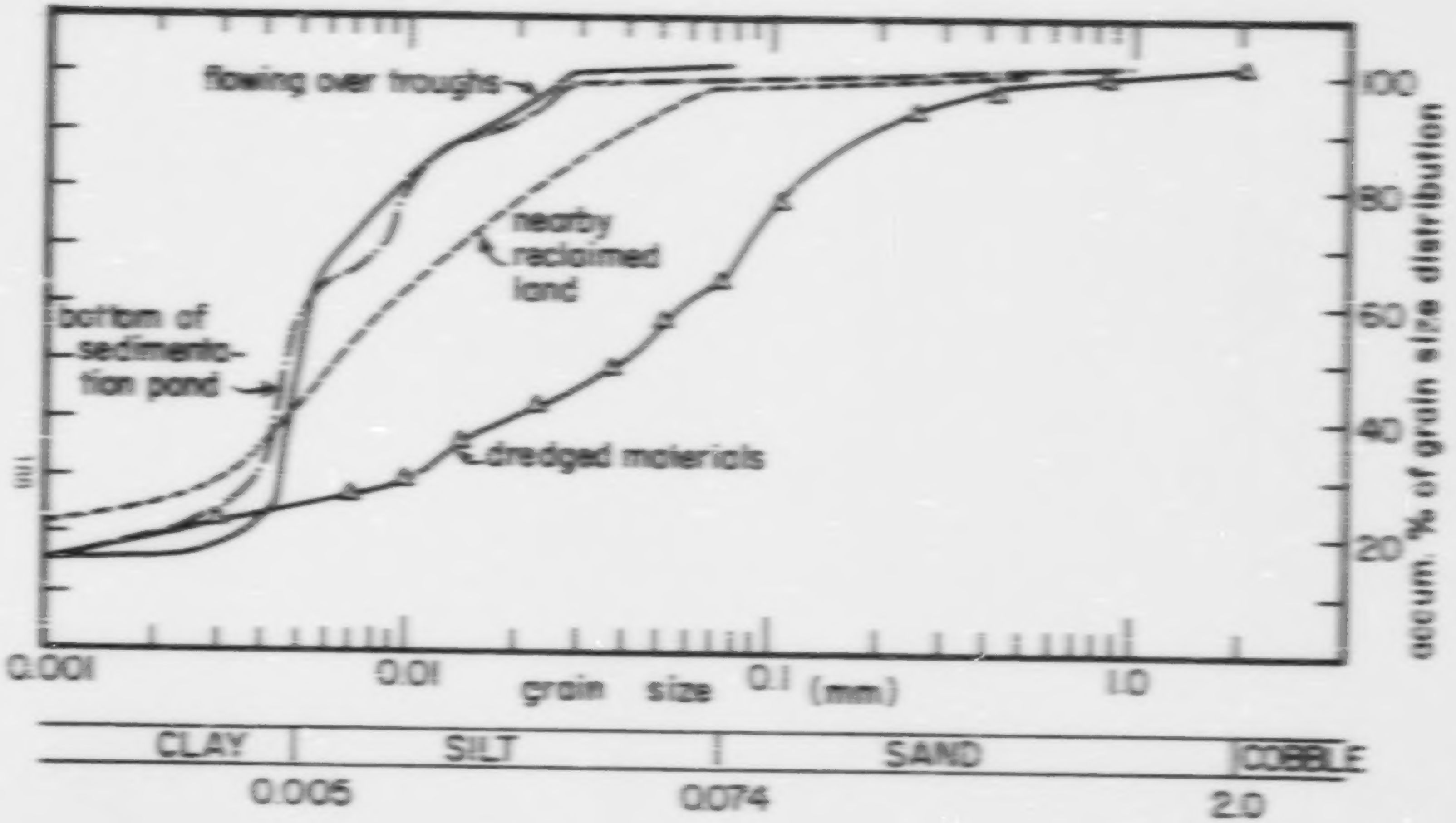


Figure 13. Grain size distributions at respective points of the trough spillway.

TABLE 3. TURBIDITY AT THE ENTRANCE AND EXIT OF THE TROUGH SPILLWAY

Measuring date	Turbidity values (average during daytime) In-flow water	 Discharged water	Effect of usage Reduced turbidity	 Removal efficiency
February 26th, 1976	35	25	10	28.6%
February 27th, 1976	33	20	13	39.4
February 28th, 1976	70	60	10	14.3
March 1st, 1976	170	150	20	11.8
March 2nd, 1976	42	35	7	16.7
March 3rd, 1976	54	38	16	29.6
March 4th, 1976	25	12	13	52.0
March 5th, 1976	32	23	9	28.1
March 6th, 1976	20	13	7	35.0
March 7th, 1976	22	20	2	9.1
March 8th, 1976	34	29	5	41.2
March 9th, 1976	16	15	1	6.3
March 10th, 1976	24	10	14	58.3
March 11th, 1976	16	15	1	6.2
March 12th, 1976	43	35	8	18.6
March 13th, 1976	35	26	9	25.7
March 14th, 1976	22	15	7	31.8
March 15th, 1976	22	13	9	40.9
March 16th, 1976	12	10	2	16.7
March 17th, 1976	25	13	12	48.0
average	36.5	28.9	7.6	20.8

Evaluation of the Trough Spillway Method

For waste water from reclamation areas consisting of more than 60% silty and clayey materials, this method could reduce the turbidity values to less than 30 ppm on the average, without using flocculants, and could operate continuously treating waste water at the rate of 5,000 m^3/hr. For land reclamation using polluted materials it would be difficult to satisfy the increased turbidity restrictions with this method without additional equipment. But this trough spillway method will be effective in combination with a rectifying sedimentation pond employing a small amount of flocculants.

AUTHOR'S POSTSCRIPT

The motivating conception from which the present report originated is an attempt to make the most effective use of all the conditions that nature offers us in the area of waste water disposal. It is important that efforts should be made without interruption in order to reduce the environmental impact that reclamation projects may cause, regardless of whether or not the soil to be reclaimed is polluted. The present report represents a small part of these efforts.

REFERENCE

Public Nuisance Countermeasuring Committee's Survey in 1974 Japan Dredging and Reclamation Engineering Association.

LEGAL AND ADMINISTRATIVE ASPECTS OF BOTTOM SEDIMENT MANAGEMENT

by
A. F. Bartsch, Director
Corvallis Environmental Research Laboratory
U. S. Environmental Protection Agency
200 S. W. 35th Street
Corvallis, Oregon 97330

INTRODUCTION

Today there is great interest in toxic substances in bottom sediments, mostly as a result of recent costly episodes in the United States and elsewhere in the world. Some episodes have tragically affected human health and welfare. Obviously, because we are confronted by these problems in both our countries, we have a kinship of concern and a united desire to deal with them in the most effective manner. To do this certainly was the intent of signatories to the May 1974 Ministerial Agreement between our countries.

In the United States we are concerned about toxic sediments in both marine and freshwaters. The marine coastlines are long and diverse, ranging from the tropics of Florida to the arctic coast of Alaska. Here and there, estuaries and bays mark areas that are susceptible to sediment deposition. There are more than 100,000 lakes ranging in size from several hectares to Lake Superior with 84,131 square kilometers, and all accumulate sediments to some extent. Major river systems have more than 418,340 kilometers of streams, many with impoundments and other segments of low velocity where sediments collect.

Sediments, and whatever toxic chemicals they may carry, enter these waters from several sources. Sediments already in place may acquire toxic substances through waste discharges or accidental spills. Apart from the sediments that come from soil erosion, principal sediments are materials discharged through pipes and materials dumped from ships and barges. Circumstances may cause some of them to become dredged spoils at a later time.

Recipient aquatic environments reflect in their accumulated sediments not only the character of the immediate surroundings but also activities taking place on the watersheds of streams that flow into them. While there is no question that even treated municipal and industrial wastes often contribute to the toxic sediments in these various settings, the materials of main consideration in this paper are those dumped from vessels. These materials are emphasized because preventive programs for wastes discharged from pipes are well established, having been continually improved over 50 years of experience, while programs to control ocean dumping and dredge spoils are still new. Traditional sources of municipal and industrial pollution are controlled under even stricter regulations in the 1972 amendments to the Federal Water Pollution Control Act (FWPCA). All point sources discharging pollutants into fresh and marine waters of the United States can legally do so only under terms of a discharge permit. By June 1976 more than 40,000 permits had been issued. In addition, all municipalities are required to provide the equivalent of secondary treatment by July 1, 1977. During the past five years, more than $5 billion of federal money was made available to cities as a national share in the cost of constructing sewage treatment works.

The United States, like many other countries, has experienced a number of episodes involving toxic substances in bottom sediments. The following three are typical examples:

--In April 1969, PCB (polychlorinated biphenyl) was first detected in the United States in oysters in Escambia Bay, Florida. Further study showed PCBs in the water, sediment, fish, blue crabs, and shrimp. The source was found to be a leak from an industrial plant 10 kilometers upstream. Since 1969, PCBs have been found in fish and shellfish from several bodies of water including the Great Lakes. The U. S. Food and Drug Administration has established temporary tolerances in food. For edible portions of fish and shellfish the level is 5 mg/kg (milligrams per kilogram), but the agency is considering lowering this level in the light of recent toxicity findings.

--In the spring of 1970, high levels of mercury were detected in fish in Lake St. Clair. To protect the public health, Canada, on the east shore, banned the sale of fish taken from the lake, and Michigan, on the west shore, did the same. These actions triggered an intense examination and search for mercury problems elsewhere and emphasized the issues of toxicity to people, bio-magnification, and ecosystem effects. During the following months bans were issued on fish and fishery products taken from waters in several other areas of the country because of mercury content.

--The most recent episode of national concern began in July 1975 when it was discovered that several workers in a chemical plant at Hopewell, Virginia, became ill and showed classic symptoms of pesticide poisoning. It was determined that they were seriously ill from massive occupational exposure to Kepone (1). As a result, the plant which was producing the Kepone was shut down a short time later. It was then discovered that during and subsequent to plant operation, Kepone had found its way into the environment, including the James River system, by way of waste effluent, seepage, and air convection. Varying amounts were found in the water, soil, and bottom sediments. Residues found ranged from 0.1 to 2.0 mg/kg in fish and shellfish, even in samples collected as far away as 64 kilometers from the source. These levels found in fin fish, crabs, and oysters caused officials to prohibit fishing in these waters. Remedial actions are still underway.

SOURCES OF POTENTIALLY TOXIC SEDIMENTS DUMPED IN OCEANS

Quantities of waste materials disposed from ships and barges increased from 1968 to 1973 and then decreased somewhat as a result of newly instituted regulatory action (Table 1). Industrial wastes and sewage sludge were dumped at sea in similar amounts up to 1974. During 1975, industrial dumping decreased markedly while sewage sludge dumping remained essentially unchanged. Over the years most of these industrial wastes have been transported to sea in vessels of 907 to 4,535 metric ton capacity to sites between 6 and 200 kilometers offshore. Greatest tonnage of dumping has been on the Atlantic Seaboard. Only industrial wastes have been dumped in the Gulf of Mexico, decreasing from 1.3 million metric tons in 1973 to 112,000 metric tons in 1975. The limited dumping in the Pacific Ocean was brought to an end by 1976.

TABLE 1. TYPES AND AMOUNTS OF MATERIALS DISPOSED IN THE OCEANS (IN METRIC TONS, APPROX.) (2, 3, 4, 5)

	Combined Total: Atlantic, Pacific, & Gulf of Mexico			
Waste Type	1975	1974	1973	1968
Industrial Waste	3,125,500	4,165,000	4,581,100	4,254,700
Sewage Sludge	4,570,900	4,544,100	4,443,300	4,060,600
Construction & Demolition Debris	359,100	698,800	883,100	520,600
Solid Waste	0	180	200	23,600
Explosives	0	0	0	13,800
Total	8,055,500	9,408,080	9,907,720	8,873,300

Industrial Wastes

The types of contaminants in industrial wastes dumped at sea vary with the industry involved. Some wastes are highly toxic; for example refinery wastes often contain cyanide, heavy metals, mercaptides, and chlorinated hydrocarbons. Data for 1973 and 1974 show dumping of mercury and cadmium off the Atlantic Coast (6). With some areas not reporting, the figures in Table 2 are minimal. In addition to mercury and cadmium, chemical plant wastes often contain many kinds of toxicants including arsenic, persistent organic compounds, and sometimes numerous other forms. New chemical products and wastes from their production create continuing challenges.

TABLE 2. MINIMUM ANNUAL INPUTS OF MERCURY AND CADIMUM
DUE TO OCEAN DUMPING OFF THE ATLANTIC COAST (6)

1974		1973	
Mercury kg.	Cadmium kg.	Mercury kg.	Cadmium kg.
1,609.7	22,348.3	2,786.8	16,407

Sewage Sludge

In the United States most sewage sludge is disposed of on land or by incineration. Only small amounts--4 million tons on a wet basis--were dumped at sea in 1968, but this amount has increased slightly each year up to 1975. Most of this sludge is dumped outside New York Harbor from origins in New York and New Jersey metropolitan areas. Because of its origin and nature, sewage sludge contains numerous residues of modern organized community living. These residues are of almost infinite variety--reflecting the food we eat, the medications we take, the cleansing agents and household chemicals we use, and the wastes from the community's business and industry tributary to the system. The range of toxicants to be expected is almost infinite also, although the concentrations may be low except in special circumstances.

Dredge Spoils

In recent years the U. S. Army Corps of Engineers has dredged an annual average of 290,519,870 cubic meters of sediments for maintenance of the nation's waterways (7). Dredging by other jurisdictions for different purposes is a much lesser activity. In 1968 and again in 1973 the tonnage of dredge spoils was about five times the tonnage of all other wastes combined (2). Moreover, it was found in 1968 that 34% of the total amount of dredge spoils was polluted based on measurements of coliform bacteria, chlorine, BOD, COD, volatile solids, oil and grease, phosphorus, nitrogen, iron, silica, color, and odor--as judged by a set of criteria still being evaluated. Dredge spoils from Lake Erie were found to contain heavy metals--cadmium, chromium, lead, and nickel--at levels deemed detrimental to aquatic life. Recently, laboratory studies seeking to evaluate the impact of open water disposal of dredged sediments have included scrutiny of metals and pesticides (7, 8).

Unfortunately, there apparently has been no search of broader scope to determine the geographical extent and frequency of toxic chemicals in marine dredge spoils. Hence, the magnitude of the toxic chemical problem can only be surmised. Nevertheless, the array of pollutants that have been found suggests that a wide variety of toxic components could be present with them.

LEGISLATION AND CONTROL PROGRAMS

In 1972, the United States passed a law as a step toward protecting the ocean, the coastal waters, and the Great Lakes. This legislative action was stimulated by a great national concern that dumping offshore was adversely affecting the marine environment. There was apprehension on the part of many people that the following vital values would be impaired:

--Provision by the Great Lakes, the coastal area, and adjacent seas of critically needed food and minerals.

--Utilization of such areas as biologically productive habitats for fish and wildlife.

--Provision of transportation, recreation, and a pleasant setting for more than 60 percent of the nation's population.

--Critical function of the oceans in maintaining the world's environment, including oxygen--carbon dioxide balance, global climate, and cycling of the planet's water.

The increasing appearance of episodes like the ones involving mercury, PCBs, Kepone, and other toxic substances that already were exacting health and economic toll added to citizen uneasiness. It was obvious that the crucial environmental values cited above could be even further impaired if pollution of the coastal waters continued to increase.

The new law, the Marine Protection, Research and Sanctuaries Act of 1972, has three main objectives: regulation of dumping, research aimed at finding ways to end all ocean dumping, and the creation of marine sanctuaries.

Procedures established under the new law to regulate dumping are now well established. The law categorically prohibits the dumping of certain materials such as high-level radioactive wastes and all biological, chemical, and radiological warfare agents. Dumping of all other wastes, with the exception of dredge spoils, is regulated by the Environmental Protection Agency (EPA). The regulations include general criteria which require that the Environmental Protection Agency consider the following factors before granting a permit for ocean dumping or for outfall discharges into the ocean:

--Need for proposed dumping or discharge.

--Effect of dumping or discharge on marine environment.

--Social and economic impacts, effects on health and welfare, fishery resources, recreation, and other values.

--Alternate disposal means available.

--Feasibility of dumping beyond the continental shelf.

Figure 1 summarizes the permit procedures in a schematic flow sequence. Only officially designated dumping sites may be used. Eleven sites in the Atlantic Ocean and Gulf of Mexico are now in use for municipal and industrial wastes. None of these wastes are dumped in the Pacific Ocean, although municipal sewage sludge enters the ocean through outfalls that are regulated under other legislation (FWPCA). Dumping site surveys are under way in several locations. They are designed to assess the impacts of dumping activities on the ecology of the dumpsite. Such derived information will provide guidance for improving regulatory procedures.

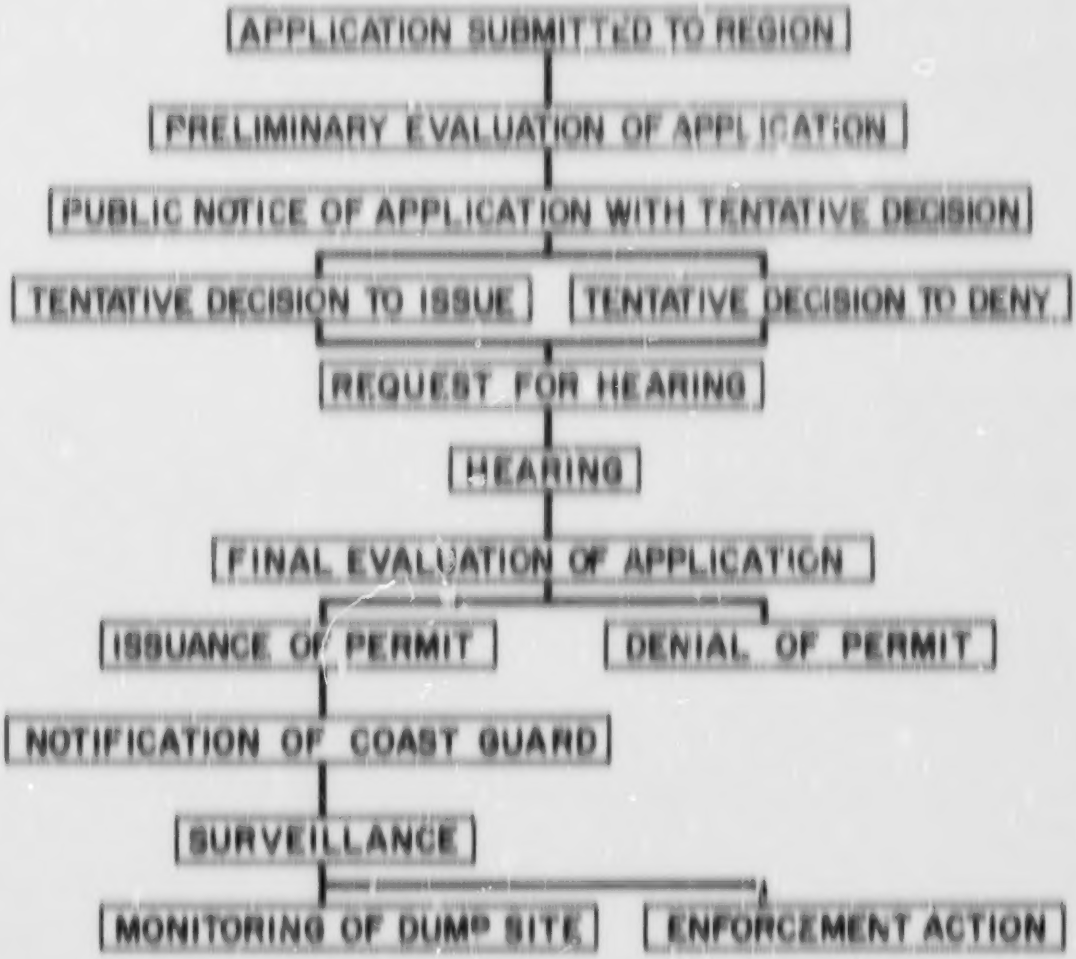

APPLICATION SUBMITTED TO REGION
PRELIMINARY EVALUATION OF APPLICATION
PUBLIC NOTICE OF APPLICATION WITH TENTATIVE DECISION
TENTATIVE DECISION TO ISSUE
TENTATIVE DECISION TO DENY
REQUEST FOR HEARING
HEARING
FINAL EVALUATION OF APPLICATION
ISSUANCE OF PERMIT
DENIAL OF PERMIT
NOTIFICATION OF COAST GUARD
SURVEILLANCE
MONITORING OF DUMP SITE
ENFORCEMENT ACTION

Figure 1. Permit procedures (6).

The Environmental Protection Agency has taken a highly restrictive approach to ocean dumping by requiring all dumpers to seek environmentally acceptable alternatives even though their wastes may meet the agency criteria for permits. As a result of this strong action, 75 former dumpers on the Atlantic Coast have stopped dumping, and 8 more were to have stopped by June of this year (9). Moreover, by a recent ruling, dumping of almost 5.5 million wet tons of sewage sludge into the New York Bright by New York and New Jersey communities is scheduled to end by December 31, 1981. Planning must begin immediately for provision of environmentally acceptable land-based disposal facilities. By the end of 1976 waste dumping in the Gulf of Mexico will be reduced to 10 percent of the 1973 figure.

The Ocean Dumping Act assigns surveillance of dumping activities to the U. S. Coast Guard. Surveillance methods include escorting or intercepting dumping vessels at the dump site by boat or aircraft, spot checking ships' logs, and using vessel riders to ascertain position and dumping rate. From April 1973 to December 1974, 983 surveillance missions were carried out, and 36 violation notifications were referred to the Environmental Protection Agency encompassing 154 apparent violations (6). Under its authority, the Environmental Protection Agency has assessed civil penalties of up to $40,000 for permit violations. There is also a provision for pursuing legal action in criminal court. During 1974, 98 permits were in force, but the number is decreasing as acceptable alternate methods of disposal are adopted.

Another aspect of control program activity relates to unexpected crises. Reactions to crisis problems like those caused by mercury, PCBs, and Kepone involve several similar stages:

--Eruption of the crisis.

--Evaluation of problem scope.

--Action to protect the public health and welfare.

--Corrective action for this episode.

--Prevention of other episodes.

The Kepone crisis may serve as an example of these reactions. Having long since passed through the first two stages, attention now centers concurrently on the remaining three. Actions to protect the public health and welfare include: (a) closure of the James River to fishing because of the Kepone levels in fin fish, oysters, and crabs; (b) monitoring Kepone levels in these organisms in the river and adjacent waters; and (c) testing citizens of the area for blood levels of Kepone.

Corrective actions underway or planned include: (a) dismantling the Kepone production plant, cleaning up the site, and disposing of the plant remnants in a state-approved landfill and (b) treatment of residual liquid wastes from production and hydroblast cleaning of the plant before dismantling. A further corrective action, step (c), relates to a problem of great magnitude--the James River sediments. The continuing contamination of fish and shellfish appears to be coming from Kepone in the bottom sediments. Removal of sediments by dredging is under consideration by the Environmental Protection Agency, the State of Virginia, and the U. S. Army Corps of Engineers. A test dredging for Kepone removal was completed recently and the results are being analyzed.

To prevent additional Kepone episodes, orders have been issued by the Environmental Protection Agency under the Federal Insecticide, Fungicide, and Rodenticide Act to stop the sale and use of this poison. To prevent other episodes of similar kind, the Environmental Protection Agency has taken steps some time ago to establish effluent standards for the following toxic pollutants: DDT (DD, DDE), aldrin/dieldrin, endrin, toxaphene, benzidine, and PCBs. Moreover, some of these agents have been de-registered for general use in the United States, and others are to be restricted for use solely by licensed, certified applicators. Such actions help to limit the opportunities for entry of these substances into bottom sediments.

Dredged material may be dumped under permits issued by the U. S. Army Corps of Engineers, but only after the permit proposal has been reviewed and approved by the Environmental Protection Agency in keeping with provisions amended to the Federal Water Pollution Control Act in 1972. In issuing such permits the Corps is required to use EPA-designated sites wherever possible as well as the disposal criteria developed by the Environmental Protection Agency as for other dumping.

New legislation recently enacted by the U. S. Congress will, if enacted into law by signature of the President, further decrease the possibilities for accumulation of toxic substances in bottom sediments. The Toxic Substances Control Act, which has been under consideration for some time, is the product of public concern over the broad dimensions of the toxicity problem--including (10):

--Magnitude of chemical use and continuing appearance of new chemical products.

--Environmental presence of toxic substances that:
are implicated in human disease*
contaminate food
cause environmental damage.

--Episodes involving:
PCBs
Vinyl Chloride
Kepone
Asbestos.

---

*Including carcinogenicity, mutagenicity, and teratogenicity.

The federal government already controls such things as food additives, pesticides, and nuclear materials. The new law will address the 1,000 or more new chemicals that enter the U. S. market each year. If passed in its present form, the proposed new law will require manufacturers to report all new chemicals three months before production, authorize the Environmental Protection Agency to screen such substances before they are marketed, order testing for potentially dangerous ones, and ban those that threaten health or the environment.

RESEARCH ON TOXIC SUBSTANCES IN BOTTOM SEDIMENTS

A provision of the Ocean Dumping Act fosters research to find ways to minimize or end all ocean dumping within five years. The results of such programs, once completed, will go a long way to mitigate the toxic aspects of such pollution. But in addition, a myriad of other research activities pursued under provisions of the Federal Water Pollution Control Act address in a broader way many aspects of the toxic substances problem--not only in marine areas, but in freshwater ones as well. The following list of specific research areas suggests the scope and significance of studies currently underway or soon to be initiated and implies the new useful information that will result from these efforts:

--Determination of acute and chronic toxic effects on marine and freshwater organisms and ecosystems caused by the following pollutants and pollution-related activities:

Complex waste mixtures.

Petrochemicals, energy-related organic solvents from offshore drilling and ocean dumping.

Onshore petroleum extraction, refining, and fossil fuel use.

Selected inorganics, complex organics, and pesticides.

Suspended particles, especially asbestos.

Kepone.

--Determination of the significance, hazards to humans, bio-magnification, persistence, transformation, and ecosystems effects of carcinogenic pollutants.

--Investigation of potential substitute pesticides to learn their chemical routes, biological effects, and degradation rates in marine and freshwater.

--Development of bioassay procedures using single species in static and flow-thru systems to estimate the ecological impact of dredge material disposal in marine and freshwater.

--Development and testing of biostatistical methods of quantitive assessment of the effects of stress on marine communities.

--Development of mathematical models of chemical equilibration of metals in seawater.

These research activities, which constitute many separate projects, are almost evenly divided between work done by government staff in the Environmental Protection Agency laboratories and grant-supported work done in universities and other research institutions. The Environmental Protection Agency laboratories most involved are the following:

Corvallis Environmental Research Laboratory, Corvallis, Oregon.

Duluth Environmental Research Laboratory, Duluth, Minnesota.

Gulf Breeze Environmental Research Laboratory, Gulf Breeze, Florida.

Narragansett Environmental Research Laboratory, Narragansett, Rhode Island.

The growing awareness of the problems of toxic substances in bottom sediments in the United States and the regulatory and research actions that have been taken as a response offer prospects for an orderly diminution of this problem. Because toxic substances easily find their way into the environment and because this problem occurs in many countries, the joint concern by the United States and Japan, the exchange of scientific information, and cooperative projects that may be undertaken will benefit all of mankind. We, therefore, must continue the effective communication established among our scientists and engineers as we move forward in responding to this challenge.

REFERENCES

1. EPA Environmental News (August 17, 1976); and EPA Kepone Fact Sheet (June 28, 1976).

2. Council on Environmental Quality, OCEAN DUMPING--A National Policy. A Report to the President, Washington, D. C. (October 1970).

3. EPA Regional Offices, Unpublished Reports--8 months of dumping activity, May to December 1973, under permits issued by Ocean Disposal Program extrapolated for 12 months to provide an annual rate (1973).

4. EPA Regional Offices, Unpublished Reports--updated information, 12 months of dumping activity (1974).

5. EPA Regional Offices, Preliminary Figures from Unpublished Reports--12 months of dumping activity (1975).

6. EPA, Ocean Dumping in the United States--1975. Third Annual Report of the Environmental Protection Agency (June 1975).

7. Chen, Kenneth Y.; Gupta, Shailendra R.; Sycip, Amancio Z.; Lu, James C. S.; Knezevic, Miroslav; and Choi, Won-Wook; Contract Report on Research Study on the Effect of Dispersion, Settling, and Resedimentation on Migration of Chemical Constituents During Open Water Disposal of Dredged Materials. Paper prepared for Joint U. S.-Japanese Conference, Corvallis, Oregon (November 17-21, 1976).

8. Fulk, R.; Gruber, D.; and Wullschleger, R.; Laboratory Study of the Release of Pesticide Materials to the Water Column during Dredging and Disposal Operations. Paper prepared for Joint U. S.-Japanese Conference, Corvallis, Oregon (November 17-21, 1976).

9. Breidenbach, Dr. Andrew W. (Assistant Administrator for Water and Hazardous Materials), Statement Before the Subcommittee on Oceans and the Atmosphere Committee on Commerce. U. S. Senate, Washington, D. C. (April 12, 1976).

10. Council on Environmental Quality, Toxic Substances (April 1971.)

HYDRAULIC DREDGING AS A LAKE RESTORATION TECHNIQUE:
PAST AND FUTURE

S.A. Peterson
Criteria and Assessment Branch
Corvallis Environmental Research Laboratory
United States Environmental Protection Agency
Corvallis, Oregon 97330
USA

ABSTRACT

Hydraulic dredging in freshwater lakes has given rise to serious environmental concerns. Examples of past dredging projects are described here and the paucity of factual data from these projects is pointed out. Lack of reliable data makes it difficult to predict the ecological effects of dredging freshwater lakes. Federal funding through the "Clean Lakes Program" will be the impetus for new lake restoration dredging projects in the United States. Potential advantages and disadvantages of dredging lakes are addressed. Lake Lansing, Michigan provides an example of the type of dredging project which may be funded. Evaluation of these projects should provide answers to many of the environmental concerns associatsd with dredging.

INTRODUCTION

In the past, "Environmentalists have pictured the dredge as the dragon in paradise, wreaking mindless destruction in valued natural resources"(1). While this view may be extreme, there are a number of potentially legitimate environmental concerns associated with dredging, especially inland lakes. These include increased turbidity, reduced light penetration, increased oxygen demand, reduced pH, alteration of water temperature patterns, release of pollutants from resuspended and newly exposed sediments, and reduced water levels, in addition to a variety of effects on the biological community. All of these may culminate in significant long range ecological modifications. However, at present there is a lack of data on many of these factors; consequently the purpose of this paper is to describe some of the problems associated with dredging nutrient enriched organic and inorganic sediments from lakes and to describe a research program to evaluate environmental concerns of future dredging activities.

Arguments in favor of dredging include improved boating and associated activities, and altered temperature patterns resulting in lower summer surface temperatures which encourage green rather than blue-green algal growth (2). One of the most positive restorative factors associated with dredging is the physical removal of the nutrients contained in the sediment.

Lake basins can be compared to reaction chambers that respond to changes taking place within them and in their watersheds. An increase in nutrient supply causes subsequent increases in productivity. The primary producers, zooplankton, and higher organisms become overabundant as a result of the increased nutrient supply. Eventually these organisms die, to be replaced by members of the same species or by different species better adapted to changing conditions. In either case the lake system continues to thrive as long as nutrients are available to the primary producers.

Nutrients are regenerated and recycled in part by both dead and living organisms in the water column; some of this organic material settles to the bottom where it undergoes further decomposition. If productivity in the euphotic zone of the lake exceeds decomposition in the aphotic zone, an oxygen deficit is incurred in the deep water with a resultant accumulation of organic debris on the bottom; the depth of the lake is reduced. Phosphorus under anaerobic, low pH conditions is highly soluble and accumulates in deep water as long as thermal stratification of the lake persists. Many lakes in advanced stages of eutrophication are shallow, however, and periodically are subject to thermal and chemical destratification by wind action. The accumulated nutrients of the deep water zone are then mixed throughout the lake, utilized by phytoplankton in the euphotic zone, and the cycle of accelerated productivity begins anew. Several investigations have referred to this partially self-sustaining mechanism of the eutrophication process (3,4,5,6).

Since eutrophic lake sediments frequently regenerate plant nutrients it is reasonable to assume that their removal from the lake would help eliminate internal nutrient loading. It should be stressed, however, that control of internal nutrient sources will be effective in reducing eutrophication only if external nutrient supplies are controlled.

PAST EXPERIENCES WITH LAKE RESTORATION VIA HYDRAULIC DREDGING

Storm Lake, Iowa

In a 1970 survey of inland lake dredging projects in the Great Lakes region, Pierce stated that, "There is no finished lake dredging project in the upper Midwest from which complete and reliable data can be obtained on the effect of lake dredging on the total lake environment"(7). He presented eight project histories of lake dredging in the upper Midwest, none of which presented conclusive evidence of benefits. An example is Storm Lake, Iowa, a shallow 1200 ha lake which has been dredged intermittently since 1940. No information is available on the total quantity of material dredged or from where it was removed. Disposal of dredged material presented problems early in the project. Dredge spoil was used to build an island in the lake and later was deposited in diked areas along the shore. Suitable material for dike construction was limited, and dike failure in 1970 forced suspension of dredging. Substantive technical information on the impact of dredging at Storm Lake is not available (7).

Trout Lake, Florida

The Florida Department of Natural Resources recognized that much of the productive fishery habitat in the state was being destroyed as a result of nutrient pollution. The mechanism was a familiar one. Sustained algal blooms precipitated detrital material to the bottom where it underwent decomposition, creating an excessive oxygen demand. This resulted in relatively non-utilized vast organic muck communities over many lake basins, where benthic fish food organisms formerly were abundant.

In 1970 and 1971, the Florida Department of Natural Resources attempted an experimental restoration at Trout Lake, a 41 ha lake near the town of Eustis (8). Approximately 31,850 m^3 of organic muck were removed, exposing 7 ha of sandy lake bottom. Lateral sloughing of the muck wall on subsequent passes of the hydraulic dredge partially recovered the exposed sandy botom. The final result was 3 ha of clean sand exposed. Over a two month period this area was further reduced to 2.5 ha as a result of additional sloughing. Cost of this project was $9,163* per hectare of bottom cleaned. The project report (8) contained no data on the characteristics of the sediment, the disposal area, or chemical characteristices of the lake environment during dredging. The only mention of biological effects indicated that in areas converted from muck to sand the diversity of the benthic population increased. It apparently now supports amphipods, prawns, clams, snails, ceratopogonids, leeches, and naiads (dragonfly, damselfly, mayfly, and caddisfly) where it formerly supported only oligochaetes and chironomids.

A subsequent report on this project is sketchy about the final outcome (9). Almost all reported changes in the lake during dredging were qualified so it was difficult to tell if the changes actually resulted from the dredging. A statement by Crumpton and Wilbur (9) indicated that increased turbidity, conductivity and total residue, decreased pH, photosynthetic activity, chlorophyll-*a*, calcium, sodium, hardness and organic nitrogen, even though temporary, could be attributed to the dredging activities. The phrase "even though temporary" is confusing since data in the appendix of their report indicated that the average concentrations of all the above parameters, plus magnesium, total phosphorus and phycophyton pigments, were greater in the two year post-dredging period than prior to, or during, dredging. It appears that a trend toward greater diversity in the benthic fish food fauna may have been realized; however, the authors reported that, "insufficient benefits were accrued to enhance the existing fish populations" (9). The long term effectiveness of the project is open to question based on the information presented.

---

* All monetary amounts quoted in this paper are given in United States dollars.

Results from the Trout Lake project produced several recommendations regarding future dredging projects in confined lake basins. Among these were that soft surface material should be removed over the entire sediment surface area prior to removing deeper sediments. Evaluations of the Florida experience further recommended that dredging begin at the highest elevations and proceed to the lowest ones. This suggests that bottom cleaning efficiency might be improved by dredging in a consecutive ring pattern beginning at the periphery of the lake basin.

Long Lake, Michigan

A hydraulic dredging project by the Michigan Department of Natural Resources from 1961 to 196[illegible] was directed toward the same objective of improving fish habitat (10). Dredging removed more than 765,000 m^3 of organic material from Long Lake in Oakland County, Michigan. The size of the lake was increased from 60 to 63 ha, and the depth from an average of 0.75 m to 2.0 m; the maximum depth increased from 2 m to 4 m. Dredge spoil was originally distributed over 6 ha of adjacent upland and marginal wetland. Problems developed immediately when solids failed to separate from the slurry, and it became apparent that additional spoil disposal area would be needed. Dredge spoil eventually covered 17 ha to a depth of 0.3 m to 1.8 m. All vegetation in the spoil area was killed. Vegetation grew on spoil after the dikes were leveled, but eight years later many of the original trees stand dead, attesting to the problems of spoil disposal. The former marshland portion resembles a wetland, but is of little value as a waterfowl habitat.

Persons using the lake now enjoy good boating and excellent bass fishing. A 1969 survey indicated that the average bass had increased in size over the pre-dredging average length by nearly 4.8 cm. Other species apparently remained approximately the same size or became slightly smaller. This might indicate a general trend in the fishery of small warm water lakes when a portion of the macrophytes are removed.

Lake Herman, South Dakota

A small portion (4.2 ha) of one bay in Lake Herman was dredged during the summers of 1970, 1971, and 1972 (Figure 1), to test the effect of hydraulically dredging inorganic silt of high nutrient concentration from the lake. Lake Herman has a surface area of 546 ha, a maximum depth of 2.4 m, and a mean depth of 1.7 m. The 145 km^2 watershed is farmed intensively for row crops and small grain. The lake is subject to periodic destratifiation due to wind action. Poor soil conservation practices have resulted in the deposition of approximately 2 m of silt over the entire lake basin.

The dredged bay area was deepened from 1.7 m to approximately 3.4 m. Some 47,860 m^3 of dredge spoil were deposited in a 3.4 ha lakeside area diked to a height of 1.5 m. Silt from the dredge slurry eventually covered the disposal area to a depth of 1.4 m. At the end of the three year project, drying had reduced the volume of this silt 50 to 60 percent.

No settling agents were added to the slurry and no attempt was made to treat the carriagewater return flow to the lake. Table 1 provides information on phosphorus concentration reductions as the dredged material moved from the sediment-water interface of the lake, through the dredge pipe, the silt deposit area, and the runoff water returned to the lake. Churchill *et al.* (11) point out that in all instances the return flow water had lower phosphorus concentrations than that in the immediate vicinity of the dredge. The significance of this is questionable, however, since phosphorus concentrations throughout the lake during the same period were more than adequate to produce the algal blooms which typically occur in Lake Herman each summer.

TABLE 1. CHANGES IN ORTHOPHOSPHORUS CONCENTRATION (mg phosphorus/l) AS DREDGE SPOIL FROM LAKE HERMAN, SOUTH DAKOTA, MOVES SEQUENTIALLY FROM THE LAKE TO THE DEPOSIT AREA, TO THE RUNOFF AREA, AND BACK TO THE LAKE (modified from reference 11)

DATE	DREDGE BAY OF LAKE	DREDGE PIPE EFFLUENT	SILT DEPOSIT AREA	DEPOSIT AREA OUTLET
7/28/70	0.29*	----	0.15	----
8/11/70	0.38	0.24	0.29	0.26
8/18/70	0.38	0.24	0.28	0.20
8/26/70	----	0.24	0.30	0.16
9/3/70	0.49	0.36	0.20	0.13
9/22/70	0.50	0.13	0.11	0.17
10/6/70	0.54	0.11	----	0.10
10/13/70	0.53	0.29	----	0.09
10/21/70	0.57	0.13	----	0.06
11/3/70	0.57	----	0.06	----
7/13/71	0.49	0.10	----	----
8/10/71	0.43	0.06	0.15	----
8/25/71	0.52	0.12	0.16	0.19
9/13/71	0.41	0.18	0	----

* At the sediment - water interface

More significant was the observation that orthophosphorus concentrations increased by approximately 300 percent (from 0.17 to ~0.5 mg P/l) shortly after dredging commenced in July, 1970. In spite of the relatively small dredging area, this magnitude of increase in phosphorus concentration was noted throughout the lake; it occurred even in the southeastern bay area which is separated from the main lake by a narrow neck of water (Figure 1). Figure 2 indicates that phosphorus concentration in the southeast bay tended to parallel that in the lake proper and approximately two months after dredging began, phosphorus concentration in the bay actually exceeded lake concentrations. Brashier *et al.* (12) attempted to assess results by comparing Lake Herman data with those collected from Lake Madison during the same period. Lake Madison is

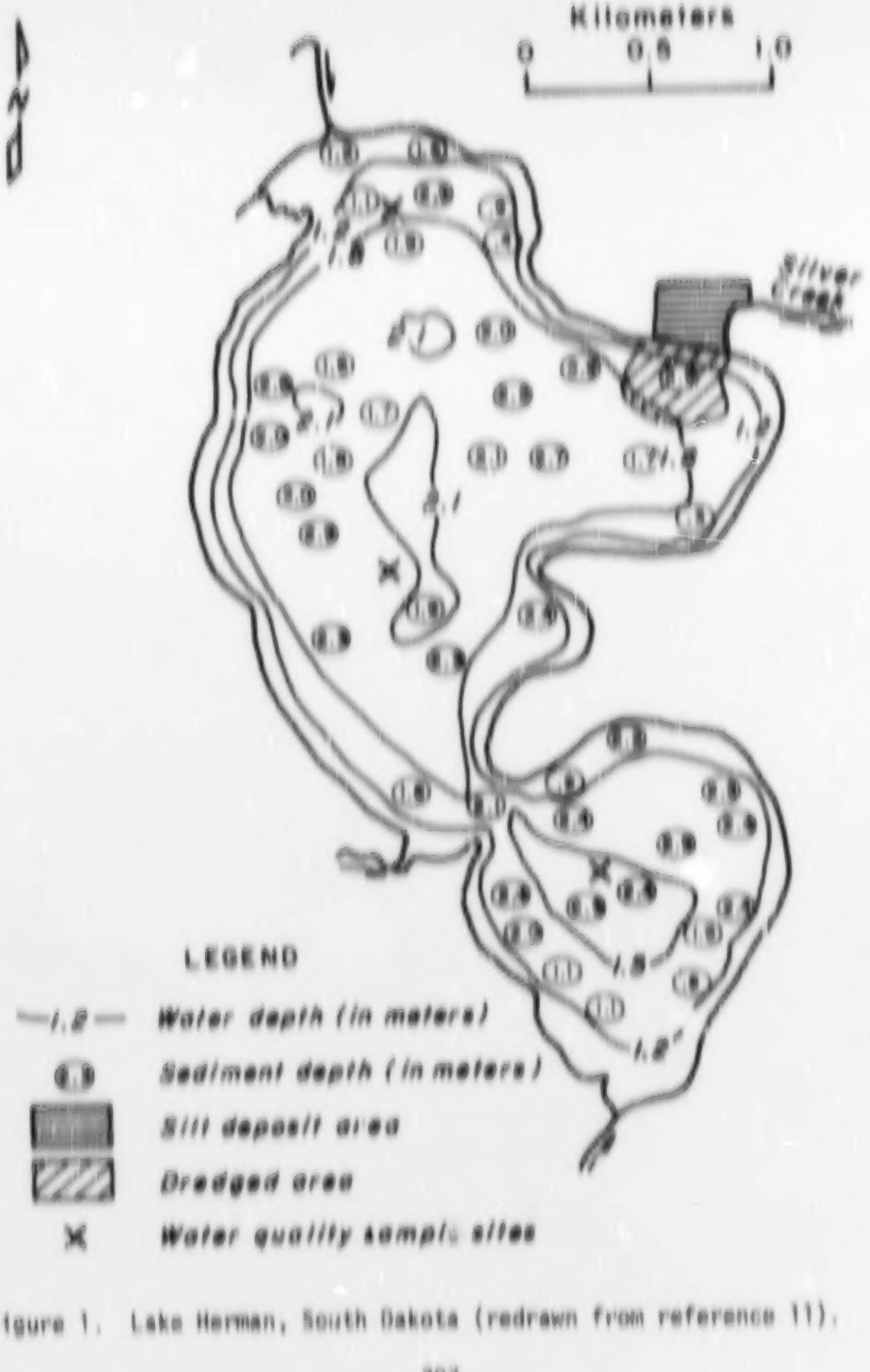

N
Kilometers
0
0.5
1.0
Silver Creek
2.1
1.5
1.2
LEGEND
—1.2— Water depth (in meters)
Sediment depth (in meters)
Silt deposit area
Dredged area
Water quality sample sites

Figure 1. Lake Herman, South Dakota (redrawn from reference 11).

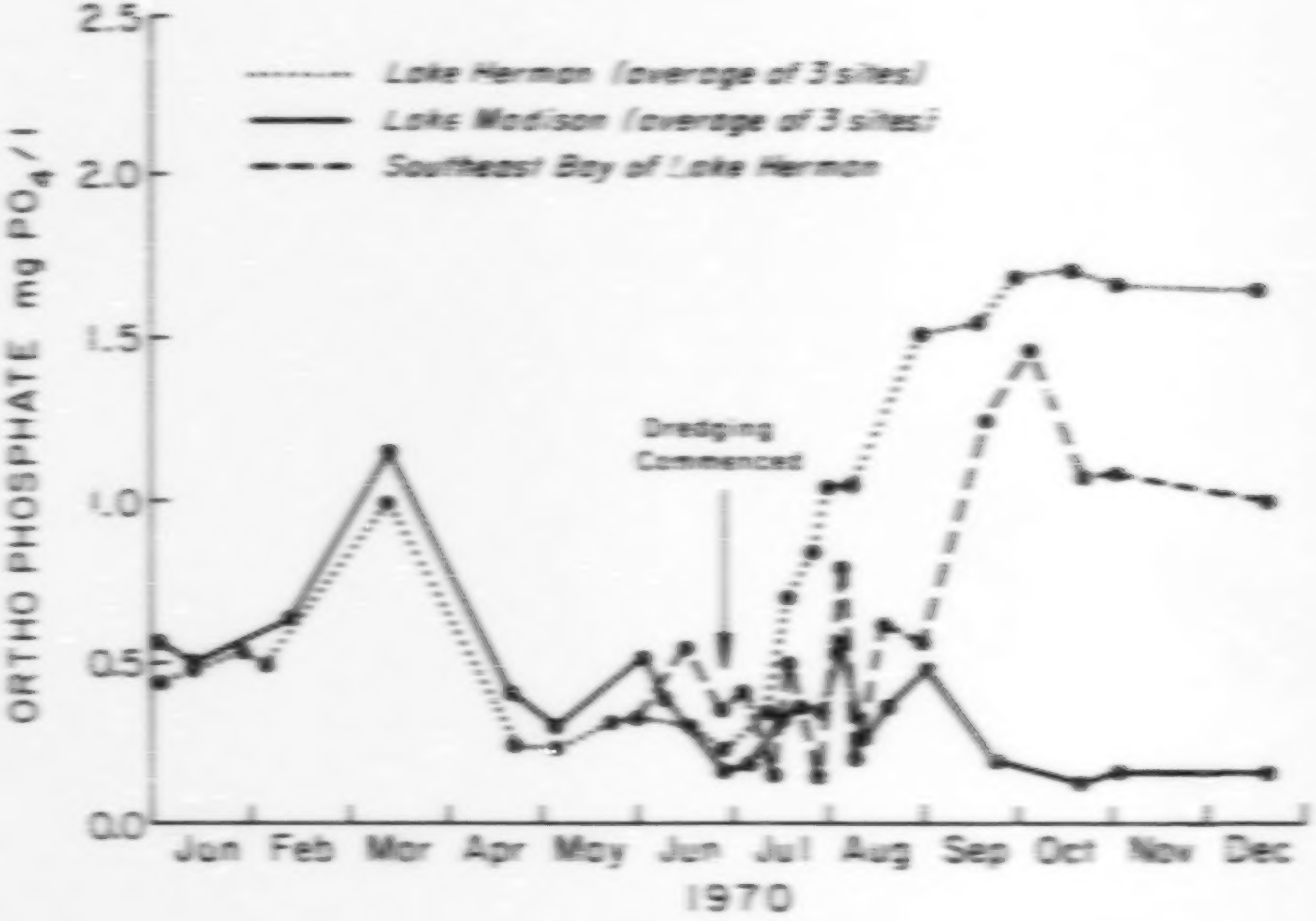


Figure 2. Orthophosphate (orthophosphate X 0.33 = phosphorus) in Lake Madison, South Dakota (modified from reference 13).

within 2-3 km of Lake Herman, but is more than twice as large (1,295 ha), has an average depth of 3.5 m, and is not subject to high silt loading. Phosphorus data in Figure 2 depict Lake Madison as a more or less typical eutrophic dimictic lake. Because of these differences, direct comparison of the two lakes is difficult. Lake Madison does, however, exemplify the typical seasonal phosphorus pattern of eutrophic, dimictic lakes in the midwestern United States, and is thus useful to show how polymictic Lake Herman differs from that pattern.

Another aspect of interest to the Lake Herman study was that phytoplankton productivity did not increase dramatically along with the increase in phosphorus concentrations (13). One explanation for this might be that Lake Herman is normally a nitrogen limited lake and increased phosphorus loading would have little impact. This is supported by data of the National Eutrophication Survey Program (14) and by Churchill *et al.* (11) who stated that during algal blooms in the lake, the nitrate-nitrogen concentration declines, sometimes to 0.0 mg/l. Another supporting factor is that nutrients other than phosphorus did not increase in the lake water during dredging. In fact, it appears that ammonia and nitrate levels in the vicinity of the dredge were actually lower than at other locations in the lake. Perhaps this is due to rapidly decreasing nitrogen concentration with increasing sediment depth as reported by Churchill *et al.* (11).

The investigators on the Lake Herman project were reluctant to attribute the increased phosphorus concentrations to dredging, since they could demonstrate no phosphorus or turbidity gradient away from the dredge. There appears to be good circumstantial evidence, however, to support a conclusion that dredging did produce the increased phosphorus levels. Table 2 from Churchill *et al.* (13) indicates there may be a trend toward lower average summer phosphorus concentrations since dredging was terminated at the end of summer 1972. Post dredging phosphorus concentration ranges appear generally to be reduced and somewhat lower on the low end of their range. Despite their reluctance to draw any conclusions about the effects of dredging on the phosphorus concentrations on Lake Herman, the authors indicated that, "there were no other noticeable environmental changes that could readily account for this dramatic increase in phosphates" (11).

TABLE 2. SUMMER PHOSPHORUS RANGES IN LAKE HERMAN, SOUTH DAKOTA, FROM 1969 to 1975 (modified from reference 13).

YEAR	ORTHO PHOSPHORUS mg/l	TOTAL PHOSPHORUS mg/l	DREDGING PHASE
1969	0.20	0.25-0.46	Prior to dredging
1970	0.17-0.50	0.17-0.99	During Dredging
1971	0.17-0.50	0.20-1.06	" "
1972	0.17-0.50	0.33-0.69	" "
1973	0.03-0.30	0.13-0.46	Following Dredging
1974	0.03-0.50	0.13-0.66	" "
1975	0.10 (through June)	0.13 (through June)	" "

Lake Trummen, Sweden

Dunst *et al.*(15) cite the court case of Reuter vs. the Wisconsin Department of Natural Resources to emphasize that a common criticism of dredging projects is a lack of concern, on the part of the dredge operators and those who commission them, for the overall environmental effects associated with dredging. One noteworthy exception to this generalization, is the project at Lake Trummen, Sweden.

By the early 1960's domestic waste discharge had deteriorated the lake to the point that filling in the basin was considered by inhabitants of the adjacent city of Växjo (16). Instead, however, a rehabilitation plan was developed by the Lake Restoration Research Team at the University of Lund. The team, consisting of limnologists, microbiologists, plant ecologists and geologists, developed a comprehensive plan to restore the lake and measure the success of their effort.

Lake Trummen has an area of 100 ha and, prior to restoration, a maximum depth of 2 m and a mean depth of 1.1 m. Studies to determine the predredge trophic status of the lake began in 1968 and continued through 1970 when the restoration project was implemented. A half meter of gyttja type sediment was dredged uniformly from the main lake basin (17). In 1971 another half meter of sediment was removed from the same area. Altogether, approximately 400,000 m^3 of sediment and an additional 200,000 m^3 of water were removed. Water content of the dredge spoil was minimized by using a specially designed intake nozzle on the hydraulic dredge. Despite the indicated removal of about 1 meter of sediment from the bottom, the lake was deepened by an average of only 40 cm. This suggests that the fine sediment may have shifted and been redistributed over the lake basin. Part of the macrophyte and gyttja dredge material was disposed of in three diked-off bays which were overgrown with macrophytes (Figure 3). Care was taken to keep the dredging of macrophyte vegetation in the lake to a minimum, consistent with Sweden's policy of nature conservation. The remainder of the dredge spoil was pumped to diked settling ponds on an old farm area from which the top soil had been removed. The return flow water was treated with aluminum sulfate to remove phosphorus and suspended solids. Total phosphorus concentration of lake water prior to restoration was about 600 μg P/l, that of the dredge slurry approximated 1 mg P/l and that returned to the lake after treatment was about 30 μg P/l. Following restoration, total phosphorus concentrations have occasionally reached levels of 70 to 110 μg P/l (18). Interstitial water orthophosphorus concentration in 1973 was 200 to 500 times lower than in 1969 (18).

The dried dredge spoil was sold to greenhouses, parks, etc. as top soil dressing for approximately $2 per cubic meter, and the proceeds helped finance construction of green belts and parks around the lake. An indication of the environmental concern evident in the Lake Trummen Project is demonstrated by the one large bay overgrown with macrophytes left intact as a waterfowl reservation (Figure 3) and an artificial island developed from dredge spoils for a waterfowl habitat.

Documentation of environmental changes associated with the Lake Trummen Project are beginning to appear in scientific literature. Bengtsson *et al.*(18) indicate that phosphorus and nitrogen have decreased drastically (Figure 4) and that the role of the sediment in recycling nutrients has been minimized. There was a general reduction in phosphorus concentration of the lake water as dredging commenced

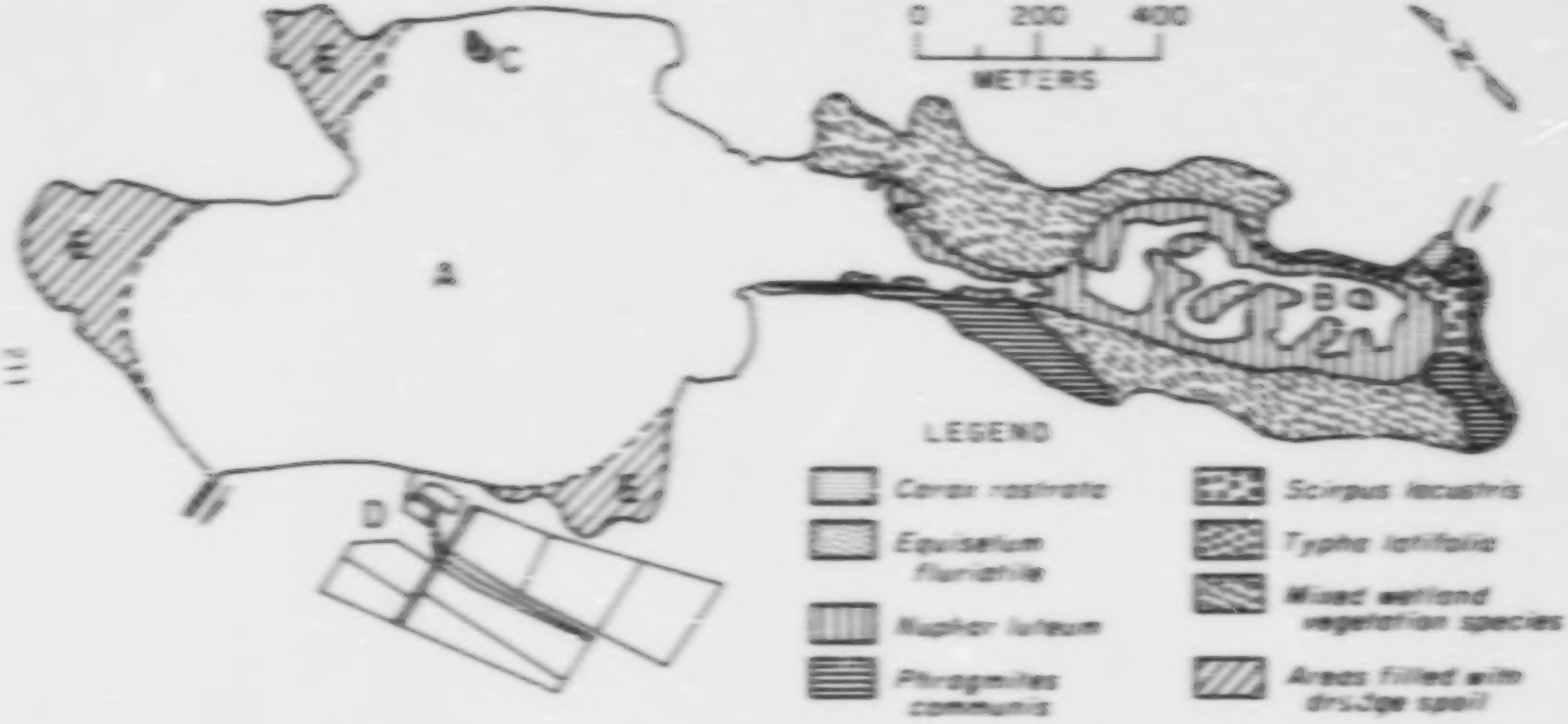

0
200
400
METERS
N
A
B
C
D
E
E
E
LEGEND
Carex rostrata
Equisetum fluviatile
Nuphar luteum
Phragmites communis
Scirpus lacustris
Typha latifolia
Mixed wetland vegetation species
Areas filled with dredge spoil

Figure 3. Lake Trummen after restoration. A. Main lake from which sediment was removed. B. Artificial island for waterfowl. D. Settling ponds and treatment plant. E. Bays filled with dredge spoil (modified from reference 17).

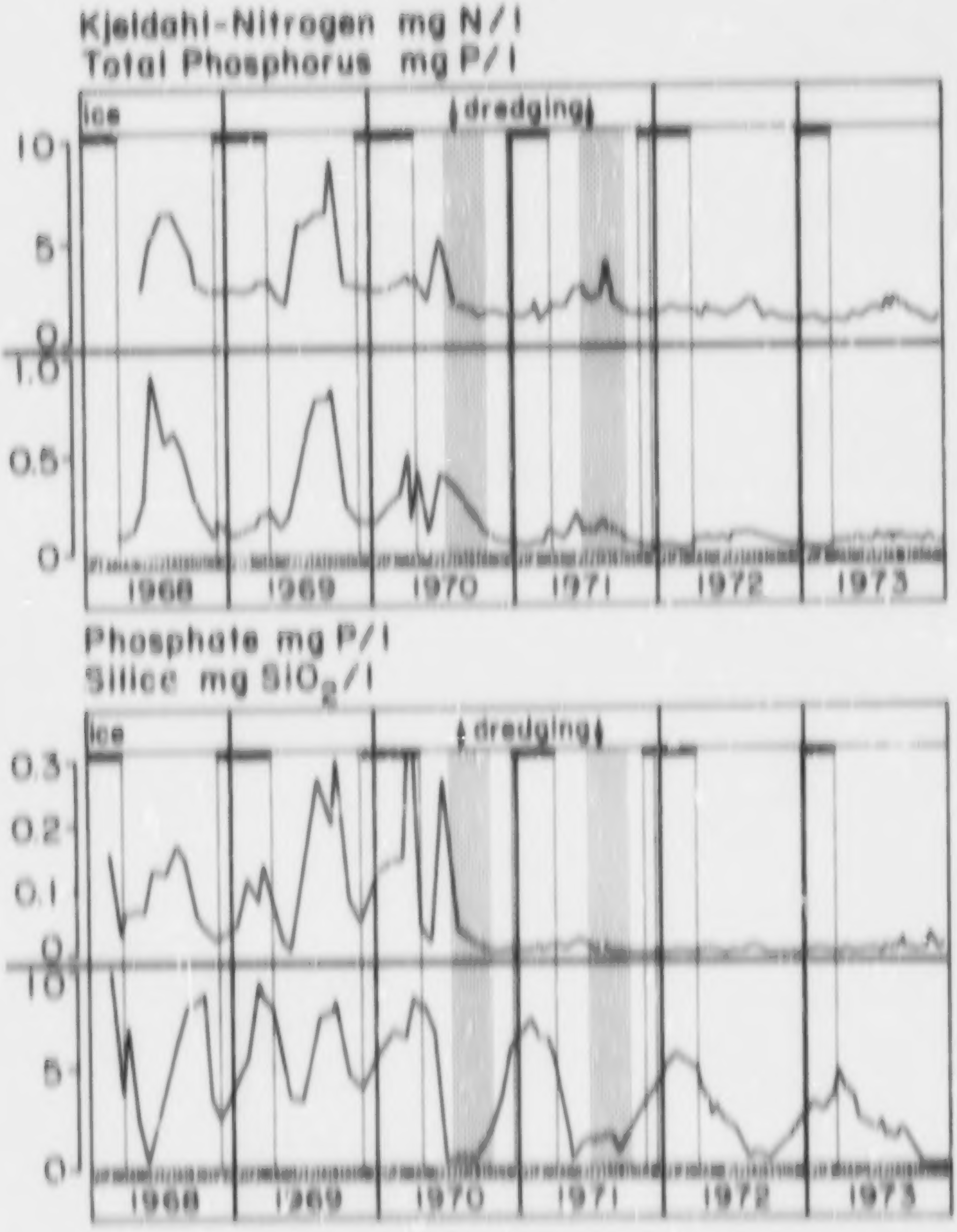


Figure 4. Kjeldahl-nitrogen, total phosphorus, phosphate phosphorus and silica in Lake Trummen (0.2 m), 1968-1973 (from reference 18).

(figure 4), just the opposite of the experience at Lake Herman, South Dakota. The total and orthophosphate as well as the Kjeldahl-nitrogen levels are significantly lower than before or during the dredging program.

Biologically, the phytoplankton diversity index (Shannon diversity index) increased from 1.8 in 1968 to 3.0 in 1973 (19). Secchi disk transparency increased from 23 to 75 cm over the same period. Prerestoration mean annual phytoplankton productivity was 370 g C/m^2 (for 1968-1969), but it declined to 225 g C/m^2 following restoration (1972-1973). In the latter case, more than 60 percent of the annual phytoplankton production was attributed to algae less than 10 μm in size. Blue-green algal biomass was drastically reduced and some species disappeared, notably *Oscillatoria agardhii*.

Disturbance of the benthic community during dredging has always concerned ecologists. Before dredging in Lake Trummen, the benthic fauna was dominated by oligochaetes and chironomids (20). A year after dredging tubificid oligochaetes and chironomids became much more numerous. Andersson *et al.* (20) indicate the most striking qualitative change in the benthos was the appearance of the clam *Anodonta cygnea*; the total number of benthic organisms changed little. They attributed this to the motility of chironomid larvae and the fact that the dominant species swarms all summer, thus probably recolonizing newly dredged areas almost immediately.

According to Carline (21), a long term project in the State of Wisconsin to determine re-population rates by benthos in dredged spring ponds exhibits varied results for similar ponds. For example, in two different ponds, after five years the chironomid populations were only 55 percent and 40 percent, respectively, of predredging numbers. In the case of amphipods, *Gammarus* increased ten fold in one case, but attained only 30 percent of predredge levels in another case. Another amphipod, *Hyalella*, was eliminated and has not reappeared. Initial reductions in benthic populations may be related to physical removal, but lack of recolonization and long term population reductions probably are more closely allied to alteration of the lake substrate.

The results to date from Lake Trummen are highly encouraging, although the data conflict somewhat with other studies. The project demonstrates that nine summer months of dredging and an expenditure of $500,000 has produced a dramatic difference in the lake and transformed it into a clean multi-use recreational facility. Changes in Lake Trummen will be followed closely through 1980 to determine the longevity of the restoration. Although the Lake Trummen project is beginning to answer many of the environmental questions associated with dredging of recreational lakes and presumably will answer many more, it will not resolve all of the questions and all of the apparent differences which have been reported in the literature. The statement by Pierce (6) about there being no finished lake dredging project with complete and reliable data on environmental effects appears to be as valid today as it was in 1970.

THE UNITED STATES CLEAN LAKES PROGRAM AND ONE EXAMPLE OF A POTENTIAL LAKE RESTORATION DREDGING PROJECT

Dredging in the United States explicitly to remove polluted sediments has not reached the scale reported in Japan. Most projects of this nature are relatively small and associated with isolated spills of toxic substances such as the one with polychlorinated biphenyl on the Duwamish River Waterway in the State of Washington (22). Most dredging in the U.S. is performed to maintain navigation lanes in estuarine waters and large rivers. Dredging to improve recreational facilities and aesthetics has been minimal. This will change as a result of Section 314 of Public Law 92-500 the amended Federal Water Pollution Control Act which states that:

A. "Each state shall prepare or establish, and submit to the Administrator for approval:

1. An identification and classification according to eutrophic condition of all publicly owned fresh water lakes in such state;

2. Procedures, processes, and methods (including land use requirements) to control sources of pollution of such lakes and;

3. Methods and procedures, in conjunction with appropriate Federal agencies, to restore the quality of such lakes.

B. The Administrator shall provide financial assistance to states in order to carry out methods and procedures approved by him under this section."

Under this law, EPA has received approximately 100 applications for financial assistance to demonstrate lake restoration techniques which will improve water quality, thereby enhancing the recreational and aesthetic attributes of the lakes. A variety of restoration techniques are being proposed, however, approximately 25 percent of those received to date, propose dredging as part of the restoration plan (Table 3). Discussion in this paper focuses on one of these potential dredging projects.

Some of the attributes as well as the environmental concerns of dredging have been mentioned previously. Another major concern in dredging fresh water lakes is the environmental effect of dredge spoil disposal. To frequently, the spoils are deposited in some adjacent wetland with little concern for environmental consequences. This may be the overriding concern associated with dredging as a lake restoration technique. Wetlands, including marshes, swamps and bogs are unique and valuable resources. They serve as habitat and breeding areas for many species of fish, waterfowl, and other wildlife, and in some cases are a valuable source of harvestable timber. Such areas may moderate extremes in water flow, serve as groundwater recharge areas, and function in natural water purification processes. They provide unique recreational

TABLE 3. LAKE RESTORATION PROJECT APPLICATIONS WHICH INCORPORATE DREDGING AS PART OF THE RESTORATION PROGRAM

LAKE	AREA hectares	AREA acres	TOTAL COST* $U.S.
Delaware Park, NY[†]	12	29	1,000,000
Blue, IA	372	918	745,000
Stafford, CA	81	200	580,500
Lake Temescal Reg Park, CA	4	10	609,450
Fenwick, WA	10	24	276,000
Gibralter, CA	121	300	10,000,000
Covell, SD	4	10	350,000
Oelwein, IA	--	--	30,000
Steinmetz, NY	1	3	78,360
Ellis Brett Pond, MA	2	5	16,000[‡]
59th Street Pond, NY	2	4	650,000
Vancouver, WA**	1052	2600	8,200,000
Long, MN	74	184	2,593,430
Liberty, WA	316	781	592,000
Silver, WA	668	1650	1,840,000
Penn, MN	15	36	245,000
Henry, WI	17	43	360,000
Noquebay, WI	871	2152	410,000
Big, WA	221	546	300,000
Lansing, MI[†]	176	435	1,600,000
Long, WA[†]	137	339	711,940
Collins Park, NY[†]	22	54	925,000
Lilly, WI[†]	--	--	546,000
			35,658,680

* Total project cost. Dredging may represent only part of this cost. Federal government pays 50%, State and Local pay 50%.

**Pilot [illegible] dredging project funded ($50,000).

[†] Projects which are funded curruntly.

[‡] Dredging portion of $1,000,000 project.

opportunities and contain many rare and endangered species. A wetland once defiled will not revert to its former level of productivity for many years, if ever.

For the above reasons, it is EPA's policy to minimize alterations in the quantity and quality of natural water flows that nourish wetlands and to protect wetlands from detrimental dredging or filling practices. Any federally funded project which anticipates an adverse effect to wetlands will require the submittal of an environmental assessment delineating the alternatives investigated and the reasons for their rejection. A cost-benefit appraisal should be included where appropriate. While the policy is strongly worded, and rightly so, it is recognized that not all wetlands are of equal value. Some are naturally more productive than others, while in some, the productivity may be optimized through various managerial practices. If, after careful evaluation of alternatives, it is decided to use wetlands as dredge disposal areas, they cannot be selected at random and for convenience. Consequently, one of the greatest problems in freshwater lake dredging is where to place the spoils in order to minimize the environmental impact, i.e., how to determine which wetlands might be filled, which ones might be modified and managed, and which ones must be preserved! The fate of many lake dredging plans will hinge on the basis of a satisfactory resolution of these problems.

Lake Lansing, Michigan

Lake Lansing provides one example of the dredging restoration projects being considered for funding under the "Clean Lakes Program" and highlights what may be the overriding environmental concern with dredging freshwater lakes--what to do with the dredge spoil.

Lake Lansing has an area of 176 ha, a drainage basin of 1029 ha, a maximum depth of 10.5 m and a mean depth of 2.7 m. Approximately 37 percent of the lake is less than 1.5 m deep and 79 percent less than 3 m deep (Figure 5). The shore line is approximately 5 km with 86 percent in private ownership and 14 percent in public ownership (1971 data). Public ownership has increased somewhat through the recent purchase of park property by the state.

Problems

The lake has long suffered from algal blooms, bacterial contamination, stunted fish populations, aquatic weed growth and the accumulation of approximately 3.5 m of bottom sediments. All have been attributed to the influx of nutrient materials from septic tank disposal systems serving permanent and summer residents around the lake (23).

According to Young *et al.*(24) the most recent blue-green algal blooms on the lake were reported in the falls of 1959 and 1962 when *Microcystis aeruginosa*, *Microcystis flos-aquae*, *Coelosphaerium Naegelianum* and *Anabaena* sp. dominated the phytoplankton. In the spring of 1960, *Ceratium hirudinella* was associated with a fish kill; that fall *Tolypothrix tenuis* was dominant in the largest bloom ever recorded on the lake.

Aquatic macrophytes generally cover the entire lake basin where the water depth does not exceed 4.5 m. The last detailed survey of aquatic macrophytes was made in 1958 (25). At that time the dominant macrophytes were *Potamogeton* spp., *Myriophyllum*, *Ceratophyllum* and *Elodea*. The lake restoration proposal compiled by John R. Snell Engineering, Inc. of Lansing, Michigan, states that macrophytes become such a nuisance after June 15 that boating is difficult. This is particularly true for sailboating, which is popular on the lake.

Chemical data for the lake are sparse; however, a general pattern of increased Kjeldahl nitrogen in the deeper water during summer stagnation reflects an increase in ammonia nitrogen levels. Total Kjeldahl nitrogen reached levels of nearly 4 mg/l during August of 1971. Increased concentrations of reduced nitrogen were not evident under ice cover (24).

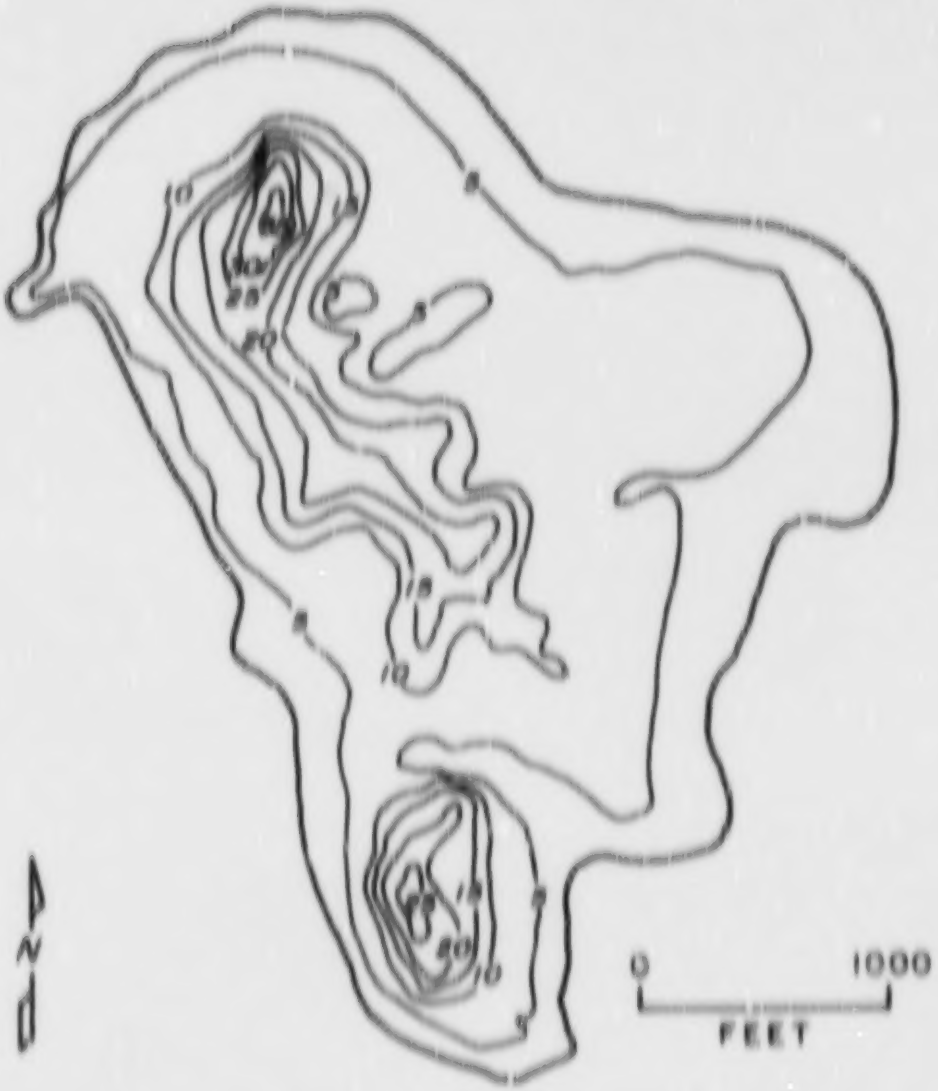


Figure 5 Contour map of Lake Lansing, Michigan, (feet X 0.3048 = meters) as originally drawn by Ball 1938 and reported by Young *et al.*, 1975. (from reference 24).

Phosphorus concentrations show a great deal of variation in Lake Lansing and there appears to be little accumulation of phosphorus in deep water during summer stagnation. Deep water samples from the lake during the winter stagnation period, however, revealed orthophosphate concentrations of approximately 150 μg P/l in March, 1972. This probably represents the maximum for the year, since the lake would ordinarily become ice free shortly after this and the concentration would be reduced by mixing with the overlying water. Biological uptake would further reduce the orthophosphorus level. The fact that nutrient levels do increase during the winter under ice suggests that the same might happen during the summer "stagnation" period.

Thermal stratification in Lake Lansing was persistent from the first week in June through most of September in 1971; however, none was indicated in two previous studies cited by Young *et al* (24). It may be that thermal stratification in Lake Lansing is intermittent, with irregular recycling of nutrients from the sediments to the lake throughout the summer. In such a case, due to more or less constant wash out, the lake may not retain high concentrations of nutrients which would tend to be released under stratified conditions. According to a draft environmental assessment of the proposed Lake Lansing project, there are no data available on the nutrient content of the lake sediments. This aspect of the project needs to be examined at the earliest possible date. The data presented by Young *et al.* (24) show that the deep water areas do become anaerobic from June through mid-September. Ordinarily these conditions would tend to cause the release of nutrients from the sediments. There was a gradual trend in this direction during the period of summer stratification in 1971, but phosphorus concentration never exceeded 80 μg P/l, not exceptionally high for a eutrophic lake.

At one time, there was some concern over the possibility of mercury toxicity as a result of dredging in Lake Lansing; however, that has been largely dispelled. The source of the mercury in this case was from atmospheric fall out, but concentrations in the sediment amounted to no more than 115 μg/kg (dry weight). Khalid *et al.* (26) found that low levels of mercury added to river sediment under controlled laboratory conditions were most readily recovered (desorbed) in their bioavailable, soluble and exchargeable forms under strongly reducing and alkaline (pH 8.0) sediment conditions. High concentrations of added mercury were most readily recovered from oxidized, acid environments (pH 5.0). Neither environment would likely be encountered in lake dredging. The same authors indicated that sediment containing high concentrations of adsorbed mercury, if oxidized, (as could happen during dredging and dredged material disposal) would most likely convert bound mercury to the bioavailable form. It was further shown, however, that the quantity released under dredging conditions would be very small relative to the total sediment mercury content. It has been pointed out that up to 50 percent of the mercury contained in dredge spoil may leave the settling pond with return flow and that this mercury may be attached to small particles constituting only 2 percent of the dry contents of the dredged sediments (27). Westermark and Ljunggren (28) have suggested that aluminum sulfate treatment might reduce the mercury content of biologically treated municipal waste effluents by more than 80 percent. Jernelov and Lann (27) suggest the same technique can be applied to dredge spoil return flow water.

The major problem with mercury, of course, is its bioaccumulation potential. The mercury content of fish in Lake Lansing has been reported to range from 0.05 to 1.01 mg/kg on a wet weight basis (29). This range represents concentrations high enough to be of concern (U.S. Food and

Drug Administration prohibits the sale of fish containing >0.5 mg/kg Hg). However, in view of the relatively small quantity of mercury contained in Lake Lansing sediment and the potential treatability of return flow water, it is thought that any additional possibility of mercury toxicity due to dredging the lake is minimal. Nevertheless, mercury in the lake and the return flow water will be monitored carefully during dredging.

As previously mentioned, the major nutrient problems in Lake Lansing have been attributed to leaching from septic tank drain field areas around the lake. Installation of a sewage interceptor system completely around the lake in 1965 alleviated much of the problem. There are few published data on the water quality of the lake following installation of the sewer system, but local residents indicate that algal blooms have decreased in recent years. At the same time, rooted macrophytes (which obtain part of their nutrient supply from the sediment) have apparently increased. These macrophyte problems, together with shallowness, which interfers with boating, have prompted the preparation of the current $1.6 million dredging proposal by the Ingham County, Michigan, Lake Lansing Lake Board. Half of the funds for this project are being provided by the Federal Government under the 314 Clean Lakes Program.

Objectives of the Lake Restoration Project

The foremost objective of this demonstration project is to remove aquatic macrophytes and organic sediment by hydraulic dredging to deepen the lake and remove the potential for resuspension of sedimented nutrients; thereby optimizing its recreational potential. A secondary objective, if funded, would be to evaluate the effectiveness of the lake restoration project in terms of improved water quality and the associated economic and social benefits realized by the local community.

Approach

The lake will be deepened selectively to 3.5 m using a cutterhead hydraulic dredge. Approximately 1.3 million m^3 of sediment will be removed from various sites which encompass approximately 80 percent of the lake basin (Figure 5). Areas to be left intact will reduce the amount of spoil to be removed and will provide habitat diversity within the lake. It is estimated that 45 percent of the sediment removed will be organic silt, 40 percent marl, and 15 percent sand. Shaping of the lake basin will result in a 10 percent slope of the bottom for a distance of 9 m from shore. From 9 to 27 m the slope will be 3.3 percent, and beyond 27 m again 10 percent to a depth of 3.5 m. Dredging will take place between April and December; an estimated three years will be required to complete the project.

Dredge spoil will be pumped to selected spoil disposal areas. The originally proposed disposal sites are shown in Figure 6 together with the area surrounding the lake considered by the consulting engineers as economically feasible for disposal. However, objections have arisen to proposed sites within the "economical pumping limit ring," because many are valuable wetland habitats (30). Project consultants do not favor disposal beyond the economical pumping limit because of the cost of pumping and returning water to the lake from outside the natural drainage basin. Return flow to the lake is necessary to maintain the water level during hydraulic dredging.

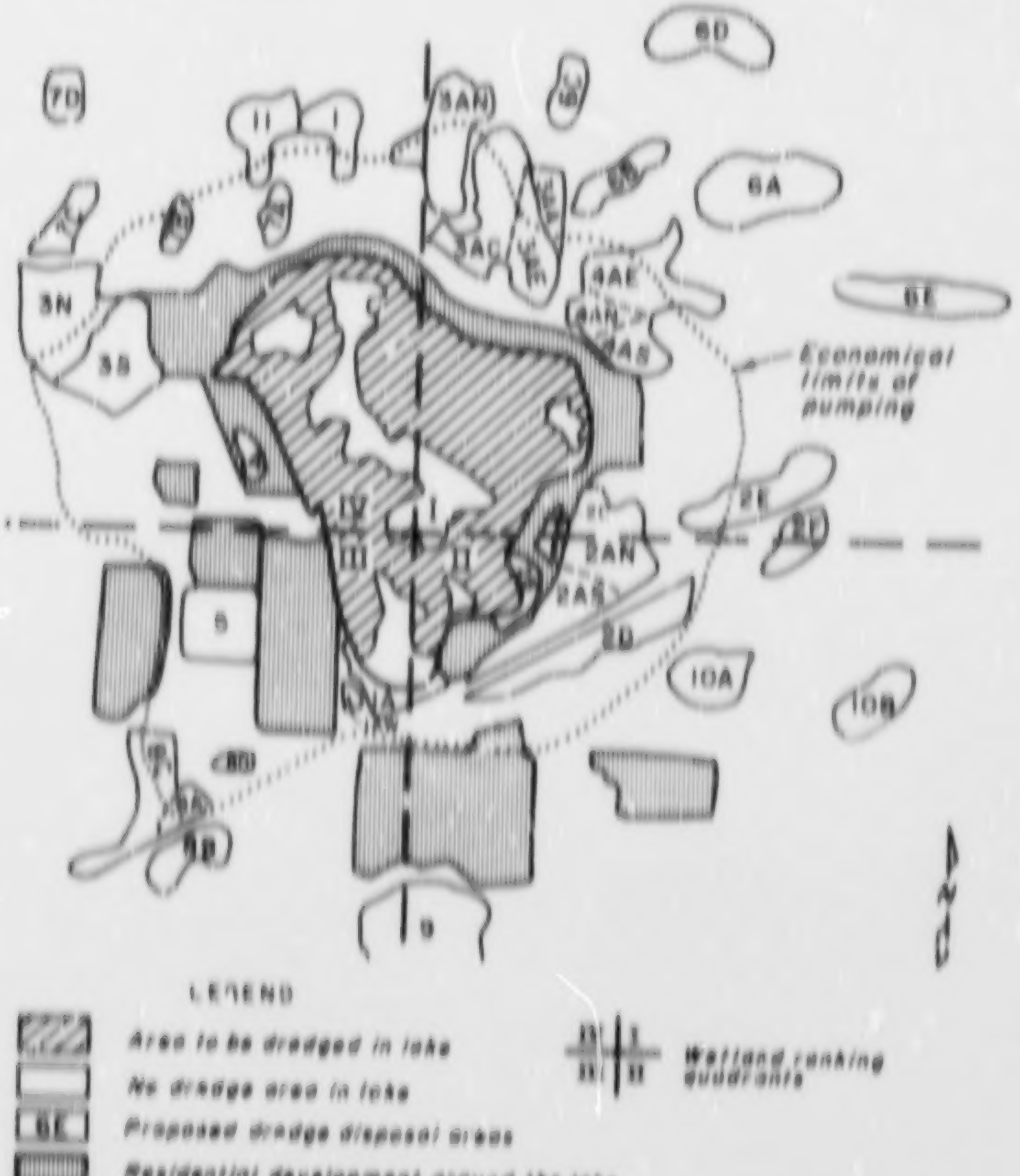

6D
7D
II
I
3AN
6A
6E
3N
3S
3AC
4AE
4AS
Economical limits of pumping
2E
2D
2AN
2AS
IV
I
III
II
5
10A
10B
8D
9
N
LEGEND
Area to be dredged in lake
No dredge area in lake
6E
Proposed dredge disposal areas
Residential development around the lake
Wetland ranking quadrants

Figure 6 Lake Lansing, Michigan showing area to be dredged, residential areas, and proposed dredge disposal sites. (modified from reference 30).

Since much of the area surrounding Lake Lansing is wetland and marsh, and since 1.3 million m^3 of dredge material requires disposal, the original proposal would result in filling nearly 70 percent of the wetlands surrounding Lake Lansing. This was unacceptable to the Michigan Department of Natural Resources, and to Federal agencies, primarily the Fish and Wildlife Service and EPA (note previously mentioned policy on protection of wetlands).

The grant for the Lansing project was funded with a proviso that alternate dredge disposal sites be selected. The consulting engineers, the Snell Environmental Group, contracted with the Department of Fisheries and Wildlife at Michigan State University to determine the relative value of wetlands bordering the lake (30). This was a positive approach to the problem; however, no new potential dredge disposal sites were added for evaluation.

The Michigan State University researchers concluded from their evaluation of the wetlands that those in quadrant I (Figure 6) were the most valuable and should be left intact as wildlife lands (30). In quadrant II, sections 2D and 9 were considered most valuable, and it was recommended they be left intact. The report concluded that all potential spoil sites in quadrants III and IV on the west side of the lake were suitable for spoil disposal. The wetland habitats were ranked by Cole and Prince according to the method of Golet and Larson (31).

The basic problem here is inadequate disposal area. To complete the project as it is currently designed, a spoil disposal area sufficient to contain 1.3 million m^3 of spoil will be required. The primary disposal sites selected by the consultants on the basis of economics and engineering priority included several areas within quadrants I and II of the Michigan State University Report (30). If the recommendation of that report concerning dredge disposal in quadrants I and II were followed, as much as 640,000 m^3 of the consultants' proposed disposal area would be eliminated from consideration. Therefore, further exploration was necessary to select alternative disposal sites.

The consultants recently developed a method of ranking the originally proposed and the subsequently selected alternate spoil disposal areas (Figure 7). These areas were ranked by a weighted system with the percentage of the final numerical index associated with each category shown in Table 4. It can be seen that the weighting system is strongly oriented toward economics and engineering. It represents an attempt to rank the potential wetland disposal sites, but ecological considerations have been downgraded. Some of the areas identified are still considered to be unfillable based on their wildlife habitat value, e.g., area number 8 in Figure 7 is the same as number 7 J (Figure 6) in the Cole and Prince report (30); an area recommended to be left intact.

Despite the lack of ecological sensitivity, the rating system may provide a focal point for discussion and final selection of dredge disposal sites. The highly biased cost factor (50 percent of rating) should be eliminated from the rating scale and disposal site selection made on the basis of minimizing adverse environmental impact. Cost will need to be considered in the final selection but should not be used as a criterion for ranking the spoil disposal areas. Final selection will have to satisfy both the grantee and EPA. If more area is required than can be reasonably agreed upon, reduction of the total amount of dredging might be required.

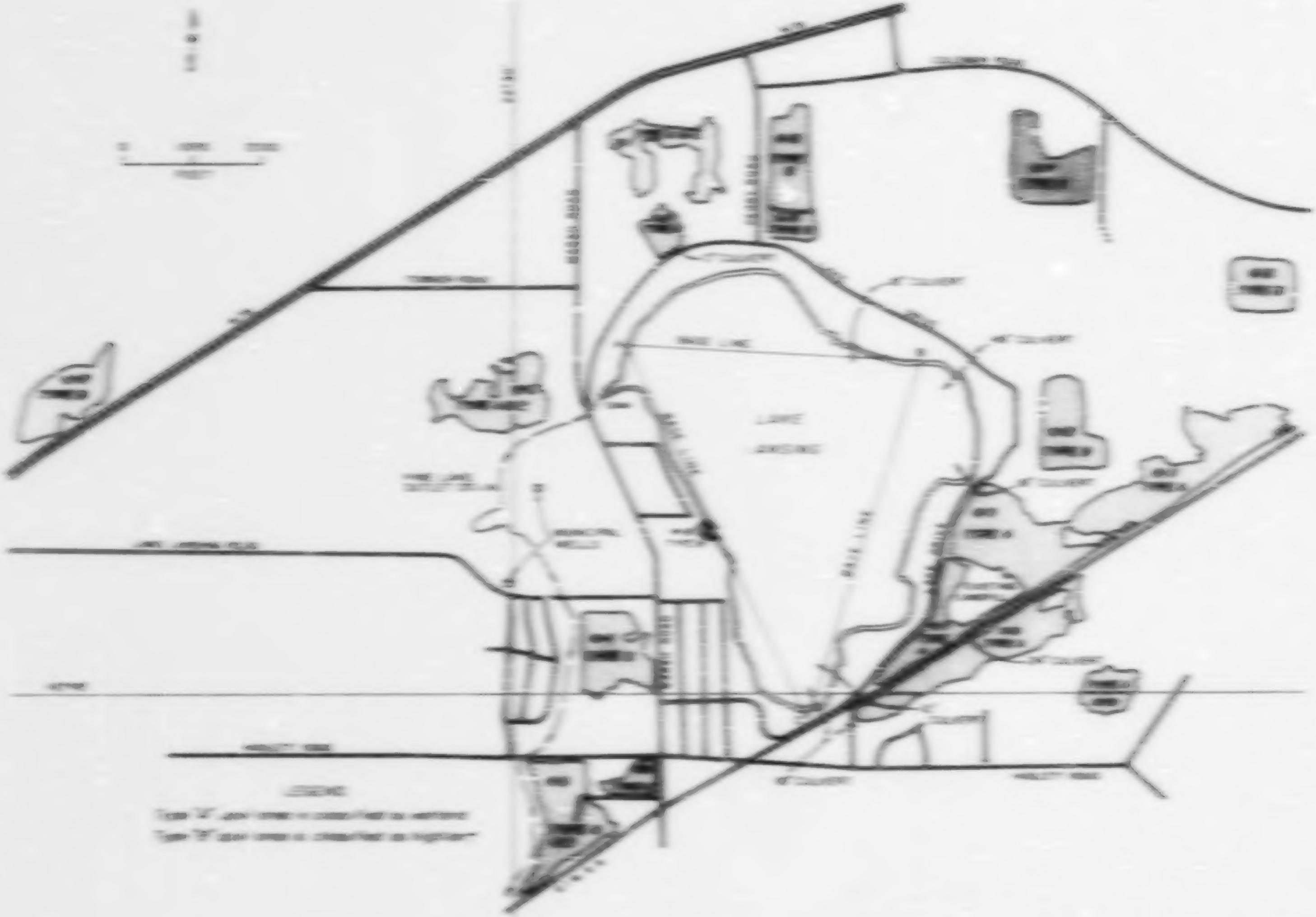

LAKE
LANSING
BASE LINE
LEGEND

Figure 7. Most recent proposed dredge disposal and alternate disposal sites for the Lake Lansing Restoration Project (from reference 32).

TABLE 4. WEIGHTED RANKING SYSTEM FOR DETERMINING IF A GIVEN AREA AROUND LAKE LANSING SHOULD BE USED AS A DREDGE DISPOSAL SITE (modified from reference 32)

	Category of Consideration	Percent of Total Rating
1)	Michigan State University wetland prioritization	10
2)	Estimated total cost to project per volume of dredge spoil	50
3)	Percent of the proposed spoil area that is true high land	10
4)	Swamp class	5
5)	Months wet/yr	5
6)	Private or public land	5
7)	Future land use	5
8)	Judgement factor	10
		100

EVALUATION PROJECT

Assuming that agreement on disposal sites is reached, research may be carried out to evaluate the results of the manipulation limnologically and socio-economically. Evaluations of this type are the primary reason for involvement of the Corvallis Environmental Research Laboratory (CERL) in the Clean Lakes Program.

A research proposal to determine the effect of dredging Lake Lansing has been submitted to EPA by Dr. C.D. McNabb of the Department of Fisheries and Wildlife at Michigan State University. According to McNabb (33) the Lake Lansing project would be evaluated in terms of changes in the following parameters:

1) mean standing crop of planktonic and filamentous algae and the maximum summer standing crop of macrophytes in the littoral zone.

2) species composition of the phytoplankton, filamentous algae and macrophyte communities.

3) rate of oxygen depletion in deep water areas of the lake.

4) maximum daily oxygen and CO_2 differences in the open water and shoreline areas of the lake during ice-free periods.

5) ratio of monovalent (Na, K) to divalent (Ca, Mg) cations in the lake.

6) availability (concentration) of carbon, nitrogen, phosphorus and silica during ice-free periods.

7) availability of toxicants (arsenic, copper, and mercury) throughout the year.

8) quantity of solids, nutrients, and toxicants discharged from the lake over the study period.

9) species composition and standing crops of fish-food organisms in zooplankton and benthos.

10) species composition, age structure, growth rates and condition factor of fish populations.

McNabb (33) has described a rational approach for evaluating this dredging project. He proposes to emphasize that which will be most directly affected, the flora and fauna of the benthic community. One of the more interesting and unique methods of analyzing the data from Lake Lansing will center around Figure 8 in which McNabb (34) suggests that recreational lakes in southern Michigan may be ranked according to a relationship between aquatic plant standing crops and the availability of essential nutrients. It has been pointed out that dredging the littoral zone of the lake to 4 m will not necessarily make this area unsuitable for aquatic plants. Post dredging analyses of reinvasion rate and species composition of the new plants will be important in evaluating dredging as a lake restoration tool.

The socio-economic analyses of this project would assess the significant changes in the economy and social structure of the community near the lake. This study would pursue two different avenues, one along the lines of a recreational travel-cost approach and the other to estimate the value of the lake to the user in terms of boating, fishing, swimming, and picnicking. The negative aspects of restoration would also be analyzed i.e., improved water quality may put undue recreational stress on the system and actually produce a detrimental effect.

The evaluation project will be realized, of course, only if the demonstration project ([illegible] lake restoration by dredging) proceeds to completion. That is dependent upon resolution of dredge spoil disposal site problems, which in turn will determine final engineering plans for the project and possible modification of the evaluation research. Therefore, the actual lake restoration at Lake Lansing could be substantially different from what is anticipated at this time. The fate of other lake restoration dredging projects in the Clean Lakes Program may be influenced significantly by the outcome of the one at Lake Lansing.

SUMMARY

--Dredging as a means of restoring the recreational potential of freshwater lakes is relatively untested.

--Past experience with dredging has produced mixed results regarding ecological effects of the projects.

--The 314 Clean Lakes Program will provide the impetus for dredging lake restoration projects in the near future; Lake Lansing Michigan is one example of the type of dredging projects being proposed.

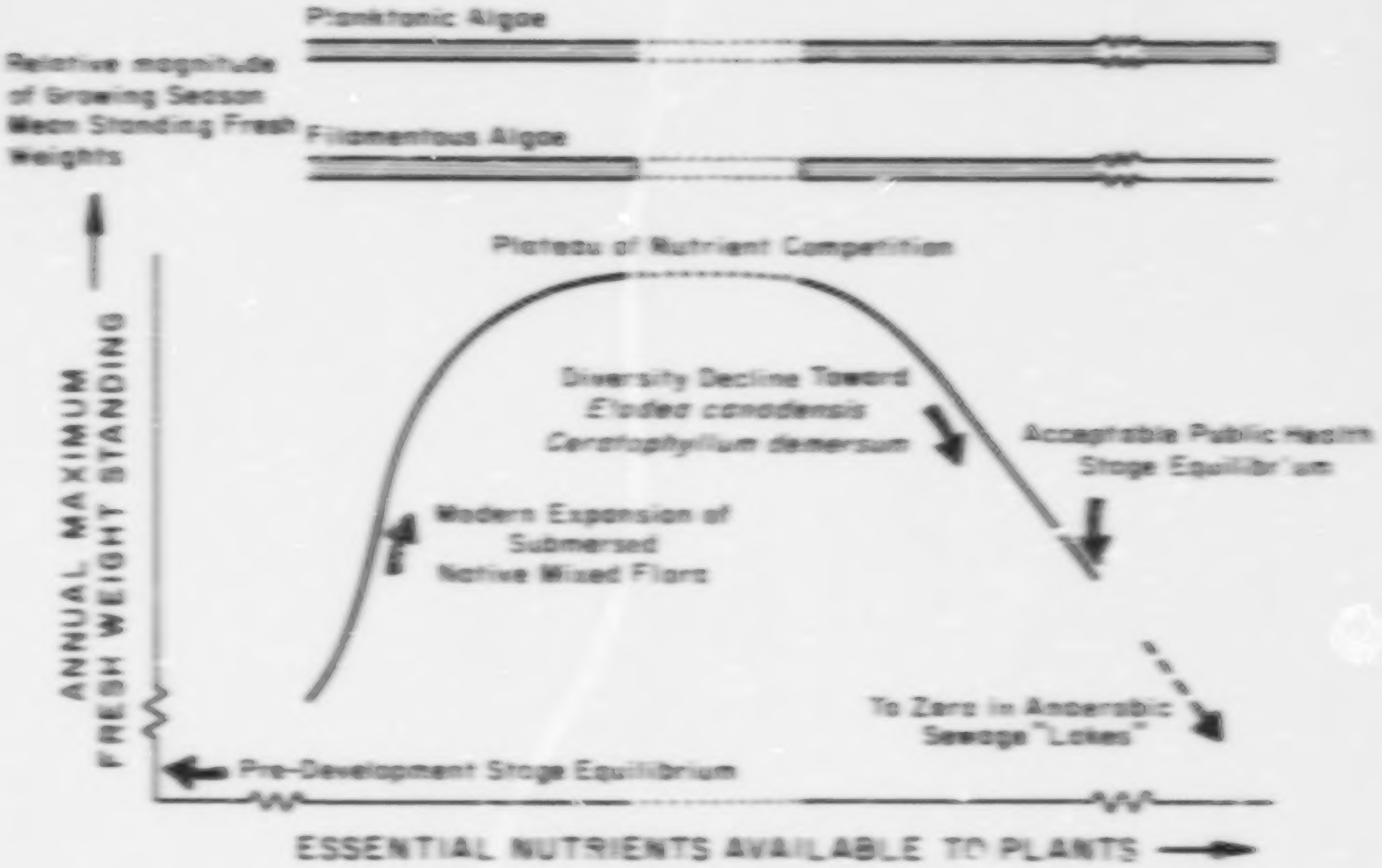

Relative magnitude of Growing Season Mean Standing Fresh Weights
Planktonic Algae
Filamentous Algae
Plateau of Nutrient Competition
Diversity Decline Toward
Elodea canadensis
Ceratophyllum demersum
Acceptable Public Health Stage Equilibrium
Modern Expansion of Submersed Native Mixed Flora
To Zero in Anaerobic Sewage "Lakes"
Pre-Development Stage Equilibrium
ANNUAL MAXIMUM FRESH WEIGHT STANDING
ESSENTIAL NUTRIENTS AVAILABLE TO PLANTS

Figure 8. Generalized relationship between standing crops of aquatic plants and increasing fertility in recreational lakes of southern Michigan (from reference 30).

--Only through proper evaluation of such projects will we be able to predict the potential for success and the environmental trade offs of future dredging projects.

ACKNOWLEDGMENTS

My sincere thanks to Russell C. Dunst, Wisconsin Department of Natural Resources, and to my CERL collegues Hal V. Kibby, Kenneth W. Malueg, Bruce A. Tichenor, Donald D. Porcella, and Charles F. Powers for their time, constructive criticism and suggestions for preparation of this paper. Thanks is also extended to Clarence D. McNabb, Jr., Michigan State University, for permission to reference his proposal to evaluate the environmental effects of dredging at Lake Lansing, Michigan.

REFERENCES

1) Turner, T. M. and Fairweather, V., "Dredging and the Environment: The Plus Side" Civil Engr. 44, 62 (1974).

2) Wetzel, R. G. "Limnology." W. B. Saunders Company. Philadelphia, Pennsylvania. 743 pp. (1975).

3) Sawyer, C. N., "Basic Concepts of Eutrophication" Jour. Water Poll. Control Fed., 38, 737 (1966).

4) Emery, R. M. *et al.*, "Delayed Recovery of a Mesotrophic Lake After Nutrient Diversion." Jour. Water Poll. Control Fed., 45, 913 (1973).

5) Larsen, D. P. *et al.* "Response of Eutrophic Shagawa Lake, Minnesota, U.S.A., to Point-Source Phosphorus Reduction." Verh. Internat. Verin. Limnol. 19, 884 (1975).

6) Haertel, L., "Ecological Factors Influencing Production of Algae in Northern Prairie Lakes." S.D. State Univ., Water Resource Inst., Completion Rep., Proj. No. A-208-SDAK (1972).

7) Pierce, N. D., "Inland Lake Dredging Evaluation." Wis. Dept. of Nat. Resources, Tech. Bull. 46, Madison (1970).

8) Wilbur, R. L., "Experimental Dredging to Convert Lake Bottom From a Biotic Muck to Productive Sand." Water Resources Bull. 10, 372 (1974).

9) Crumpton, J. E. and Wilbur, R. L., "Habitat Manipulation." Dingell-Johnson Job Completion Report, Proj. No. F-26-5. Florida Game and Fresh Water Fish Comm. (1974).

10) Spitler, F. J., "Dredging Along Lake Michigan to Improve Boating and Fishing." Mich. Dept. Natural Resour. Tech. Bull. 73-17 (1973).

11) Churchill, C. L., *et al.*, "Silt Removal From a Lake Bottom." EPA Ecol. Res. Ser. 660/3-74-017 (1975).

12) Brashier, C. K., *et al.*, "Effect of Silt Removal in Prairie Lake." EPA-Ecol. Res. Ser. R3-73-037 (1973).

13) Churchill, C. L. *et al.*, "Evaluation of a Recreational Lake Rehabilitation Project." OWRR Comp. Report No. B-028-SDAK. Water Resources Inst., South Dakota State University, Brookings (1975).

14) National Eutrophication Survey. "Preliminary Report on Lake Herman, Lake County, South Dakota, EPA Region VIII," Corvallis Environmental Research Laboratory, Corvallis, Oregon. 7pp mimeo (1976).

15) Dunst, R. C., *et al.*, "Survey of Lake Rehabilitation Techniques and Experiences." Wis. Dept. Nat. Resources Tech. Bull. 75, Madison (1974).

16) Björk, S., *et al.*, "The Lake Trummen Restoration Project: A Presentation." The Lake Restoration Researchers Team, Univ. of Lund, Sweden. 36p mimeo. (1971).

17) Björk, S., "European Lake Rehabilitation Activities." Plenary lecture at the Conference on Lake Protection and Management, Madison, Wis. Oct. 21-23, 23 p. mimeo (1974).

18) Bengtsson, L., *et al.*, "Lake Trummen Restoration Project I. Water and Sediment Chemistry." Verh. Internat. Verein. Limnol. 19, 1080 (1975).

19) Cronberg, G., *et al.*, "Lake Trummen Restoration Project II. Bacteria, Phytoplankton, and Phytoplankton Productivity." Verh. Internat. Verein. Limnol. 19, 1088 (1975).

20) Andersson, G., *et al.*, "Lake Trummen Restoration Project III. Zooplankton, Macrobenthos and Fish." Verh. Internat. Verein. Limnol. 19, 1097 (1975).

21) Carline, R. F., Project Leader, Wisconsin Department of Natural Resources, P. O. Box 203, Waupaca, Wisconsin, USA 54981.

22) Pavlou, S. P., *et al.*, "PCB Monitoring in the Duwamish River: A Study of Their Release Induced by the Dredging Activities in Slip-1." University of Washington, Department of Oceanography, Special Report No. 66, Ref. No. M76-46, Seattle (1976).

23) Kettele, M. J. and Uttormark, P. D., 'Problem Lakes in the United States." University of Wisconsin Water Resources Center Tech. Report 16010 EHR 12/71, Madison (1971).

24) Young, T. C., et al., "Project Completion Report Predredging Analysis of Lake Lansing, Michigan." Michigan State University, Institute of Water Research Tech. Report No. 43 (1975).

25) Roelofs, E. W., "The Effect of Weed Removal on Fish and Fishing in Lake Lansing." Dept. of Fisheries and Wildlife, Michigan State University, East Lansing. 10pp (1958).

26) Khalid, R. A., et al., "Sorption and Release of Mercury by Mississippi River Sediment as Affected by pH and Redox Potential." Paper presented at 15th Annual Hanford Life Sciences Symposium, Richland, Washington Sept. 29 - Oct. 1, ERDA Symp. Series (1975).

27) Jernelov, A. and Lann, H., "Studies in Sweden on Feasibility of Some Methods for Restoration of Mercury--Contaminated Bodies of Water." Environmental Science and Technology 7, 712 (1973).

28) Westermark, T. and Ljunggren, K., "Report to the Swedish Applied Research Council." No. 4952 (1968).

29) D'Itri, F., et al., "An Estimation of the Total Mercury Content in Some Lake Lansing Fish and Sediments." Reprint Presented to the Lake Lansing Lake Board Meeting, Meridian Township, Michigan. (1971).

30) Cole, R. A. and Prince, H. H., "Fish and Wildlife Values of Wetlands Bordering Lake Lansing Proposed as Potential Spoil Disposal Sites." Dept. of Fisheries and Wildlife, Michigan State University, East Lansing. Report prepared for Snell Environmental Group Consulting Engineers in support of Lake Lansing Restoration Project EPA No. 66405, April 26, (1976).

31) Golet, F. C. and Larson, J. S., "Classification of Freshwater Wetlands in the Glaciated Northeast." Bur. Sport Fisheries and Wildlife, Research Publication No. 116. 56pp. (1974).

32) Personal Communication. Letter from Snell Environmental Group, Lansing, Michigan, dated August 27, 1976.

33) McNabb, C. D., "Evaluation of Dredging as a Lake Restoration Technique." Michigan State University Dept. of Fisheries and Wildlife. A research proposal submitted to EPA for Federal assistance. (1976).

34) McNabb, C. D., "Aquatic Plant Problems in Recreational Lakes of Southern Michigan. Michigan Department of Natural Resources, Lansing, 53pp. (1975).

INTERCHANGE OF NUTRIENTS AND METALS BETWEEN SEDIMENTS AND WATER DURING DREDGED MATERIAL DISPOSAL IN COASTAL WATERS

by

D. J. Baumgartner, D. W. Schults, S. E. Ingle and D. T. Specht

Marine and Freshwater Ecology Branch
Corvallis Environmental Research Laboratory
U. S. Environmental Protection Agency
Corvallis, Oregon 97330

ABSTRACT

Conventional barge dumping of over [illegible],000 m^3 of channel sediment was arranged by the Corp[illegible]f Engineers at a controlled experimental dump site in Elliott Bay, Puget Sound to study the fate and effects of metals, polychlorinated biphenyls (PCBs), and nutrients in the dredged materials. Periodic sampling was conducted before, during, and at several intervals after dumping to determine the distribution and uptake of materials as well as other biological effects resulting from the dumping. The objective was to provide guidance to the regulatory agencies regarding the effects of disposal of material containing measurable levels of pollutants in "open water" disposal sites. Incidental to this purpose, research was undertaken to improve analytical techniques, handling of samples and understanding of chemical pollutant mobilization between water and sediment as it affects the marine environment.

The purpose of this paper is to present an overview and some partial results of this unusual project and to describe methods employed.

Perturbations in some water quality parameters were considerable but short term during the dumping operation. The concentration of chemical species in the water were, however, below the values recommended as maximum concentrations by the Committee on Water Quality Criteria (U. S. Environmental Protection Agency, 1973). Partial results of sediment analyses indicate that there has not been substantial chemical alteration of the disposal site environment.

INTRODUCTION

Both the United States Environmental Protection Agency (EPA) and the Corps of Engineers (COE) are seeking information about the environmental impact of dredging and disposal operations so that these operations will be technically satisfactory, economically feasible and yet protect the environment. Part of the COE research is to evaluate the environmental effects of open water disposal of dredge material as authorized under the 1970 River and Harbor Act. Similarly, the EPA under the Issuance of Permits for Dredge and Fill Material (Section 404 of Public Law 92-500) needs additional information to establish guidelines and procedures on open water disposal. To learn more about environmental effects associated with open water disposal of dredged material, the COE initiated the Duwamish Waterway Project in Puget Sound, Washington. The dredging of the Duwamish Waterway with subsequent marine "open water" disposal of the dredge material into Elliott Bay on the river delta, offered an opportunity to observe environmental effects principally from heavy metals, PCBs and nutrients. The objectives of the field investigation are to:

a. Document the release of chemical species from the dredged material to the water column during the following disposal operations.

b. Measure uptake, if any, of heavy metals and PCBs by important species of demersal fish and shellfish.

c. Determine the effect of dredged material disposal on benthic and demersal faunal abundance and distribution and the rate and extent of benthic recolonization in the area of the dredged material deposit.

The role of EPA in the cooperative study is twofold: (a) to provide data on routine analyses of chemical constituents, and (b) to conduct research on the behavior and effects of certain chemicals as particulates move from the barge and become incorporated into the sea bed. Both field and laboratory investigations are being performed to define the extent of release and movement of chemical species. Laboratory evaluations include long and short term release of heavy metals from deposited material to seawater; response of a marine test alga in seawater before and after mixing the seawater with dredged materials; and, in the laboratory, the response of benthic animals to deposited material. Much of the research is presently in progress; therefore, this report will present initial results of heavy metals and nutrients for the field investigations before, during and after the disposal operation and the results of the laboratory algal assay.

The National Marine Fisheries Service is documenting the biological uptake of heavy metals and PCBs while Shoreline Community College is documenting the rate and extent of benthic recolonization at the disposal site.

The analyses of PCBs in sediment and seawater are being conducted by Dr. Spyros Pavlou at the University of Washington Oceanography Department.

SITE DESCRIPTION

The study area is located in the southeast section of Elliott Bay in Puget Sound, Washington (Figure 1). Between January 27 and March 6, 1976, approximately 114,000 m^3 of sediment were removed from the mid channel of the Duwamish River and deposited in Elliott Bay near the river mouth at a depth of about 60 m. Dredging was done on a 1.9 km section of river (river km 6.3 to 8.2) using a clamshell dredge and barge transportation to the disposal site. The dredged river sediments were characterized as sandy silt whereas the bay sediments are typically sand.*

PARAMETERS MEASURED

The chemical and physical parameters that were measured are listed in Table 1. The main emphasis of the study was on heavy metals, PCBs and nutrients. Techniques used to preserve and analyze the samples are outlined in Table 2.

SAMPLING PROCEDURE

Sediment samples were taken by gravity cores in parallel and by Van Veen grab for bulk analyses. Water samples for nutrients and metals were pumped from three depths using plastic hose; samplers fabricated from stainless steel beer barrels were used for collecting PCB samples. Water for PCB analysis was filtered on board the Research Vessel Hoh (R/V Hoh) using glass fiber filters in stainless steel holders.

Sediment cores were obtained from the Research Vessel Streeter. One core was processed on board for subsequent PCB analysis. A second core was held for metal, grain size, and nutrient analysis.

SAMPLING SCHEDULE

The sampling was divided into three main periods: background, disposal period, and post-disposal period. During the background sampling, 19

---

*During the oral presentation 14 color slides were shown to aid in visualizing the equipment employed for sampling and for handling the dredge material. They have not been reproduced for this report.

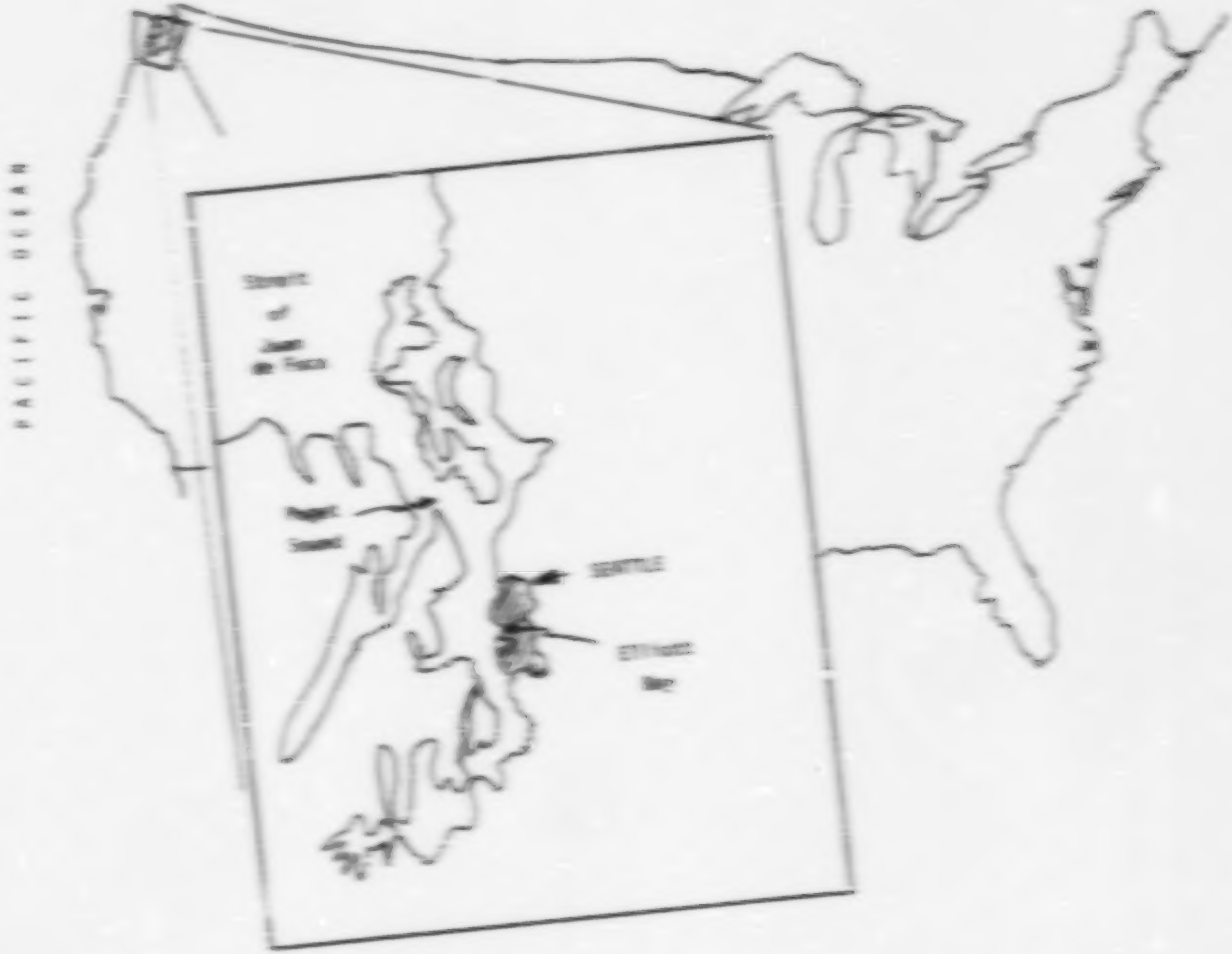

PACIFIC OCEAN
Strait of Juan de Fuca
Puget Sound
SEATTLE
Elliott Bay

Figure 1. General map of study area

TABLE 1. CHEMICAL PARAMETERS

I. Water

pH
NH_3
$NO_3 + NO_2$
OP
dissolved Cd, Cr, Pb, As, Fe, Mn
total Hg
particulate metals
suspended solids
PCB
oil/grease

II. Sediment

Total	Interstitial
pH	dissolved Cd, Cr, Pb, As, Hg, Mn
Cd, Cr, Pb, As, Fe, Mn, Hg	NH_3
organic C	$NO_3 + NO_2$
Total and free sulfides	OP
Eh	PCB
particle size	
PCB	
oil/grease	
moisture	

III. Elutriate test

NH_3
$NO_3 + NO_2$
OP
dissolved Cd, Cr, Pb, As, Hg, Mn, Fe
PCB

TABLE 2. SUMMARY OF PRESERVATION AND ANALYTICAL METHODS

Parameter	Preservation	Method of Measurement
Sediment moisture	-----	dry at 70°C
Soluble Cd, Cr, Pb, Mn, Fe in seawater	1% HNO_3	atomic absorption spectrophotometer (AA)
Hg in seawater and sediments	1% HNO_3+16ppm Au^{+++}	digestion in aqua regia, cold vapor AA analyses
Cd, Cr, Pb, Mn, Fe in sediments	freeze dried	HNO_3/HF digestion, AA analyses
NH_3-N	40 mg/l $HgCl_2$	distillation, automated indophenol blue
NO_2-NO_3	40 mg/l $HgCl_2$	automated cadmium reduction followed by diazotization
Ortho P	40 mg/l $HgCl_2$	Murphy-Riley
Soluble As in seawater	1% HNO_3	arsine generation, AA analyses
As in sediment	freeze dried	HNO_3 digestion/plasma emission spectrophotometer
soluble sulfide in sediment	frozen in absence of air	cadmium nitrate titration using sulfide electrode
total sulfide in sediment	frozen in absence of air	acid generation, cadmium nitrate titration using sulfide electrode
Eh	-----	Pt-calomel electrode
particle size distribution	air dried	hydrometer technique
suspended solids in seawater	-----	0.45μ membrane filter, dry at 70°C
organic carbon	freeze dried	O.I.C. carbon analyzer
PCB	hexane	gas chromatography

river sediment cores (length to 0.6 m) were obtained with a Benthos Gravity Corer from river sm 6.7 to 8.2. Likewise, cores were taken from 16 stations at the disposal site and four reference stations (Figure 2). Analyses were done on the total core and interstitial water. To evaluate the pollution potential of the river sediment, additional tests were performed including elutriate test, algal assay of elutriate test water and benthic bioassay using the river sediment. The purpose of this background sampling was to predict what chemical species might be solubilized and of concern during and after disposal of the river sediment to Elliott Bay, and to document any algal and benthic response to the dredged material.

Sampling during the disposal period consisted of collecting water during three consecutive disposal operations on two days. Each disposal operation consisted of dumping two barges with 380-535 cubic meters of river sediment each. Water was collected from two vessels, one at the disposal site center (R/V *Hoh*) and the other about 60 m downwind (northeast) from the site center (R/V *Streeter*). Each vessel collected water at the same time from three depths, 1 m below the surface, mid depth and 1 m above the sediments. The sampling frequency was: -15, 0, 5, 10, 15, 25, 45, 75, and 120 minutes after the dump. Water samples were also collected at the reference stations at the beginning and at the end of each day's monitoring. The main objective of monitoring the disposal period was to identify the release of chemical species from the dredged material to the water column.

The activities during the post-disposal period consisted of sampling the sediments and the water column at the disposal site and the reference stations. Sediments were collected 1 week, and 1 and 3 months after the dumping was completed. The upper 10 cm and the lower portion of the sediment core were analyzed separately. The water was sampled at the same three depths as during the disposal period. The purpose of the post-disposal sampling was to determine the long term effects of dredged material disposal on the migration of chemical species in the sediment and to the water column.

RESULTS

All the sediment and water samples have not been analyzed, but the initial data indicate some distinct differences between the bay sediment and dredged river sediment. Some of these differences can be seen in Table 3. Cadmium was not detected and lead was lower in the river sediment than in Elliott Bay sediment prior to dumping, so those two metals were not measured during the disposal and post-disposal periods. Mercury levels in the river sediment were similar to those in the bay sediment but it was monitored because of its significance in the environment. Arsenic and chromium were at sufficiently high levels in the river sediment to be of concern. Ammonia was about an order of magnitude higher in the river sediments than in the bay sediments and at a level that could produce an impact to the immediate disposal area.

From water column suspended solids data (Figure 3) obtained during the disposal operation the dredged material appeared to settle through the water column as a slug rather than in an extensively dispersed condition. This tended to transport much of the soluble chemical species in the dredged

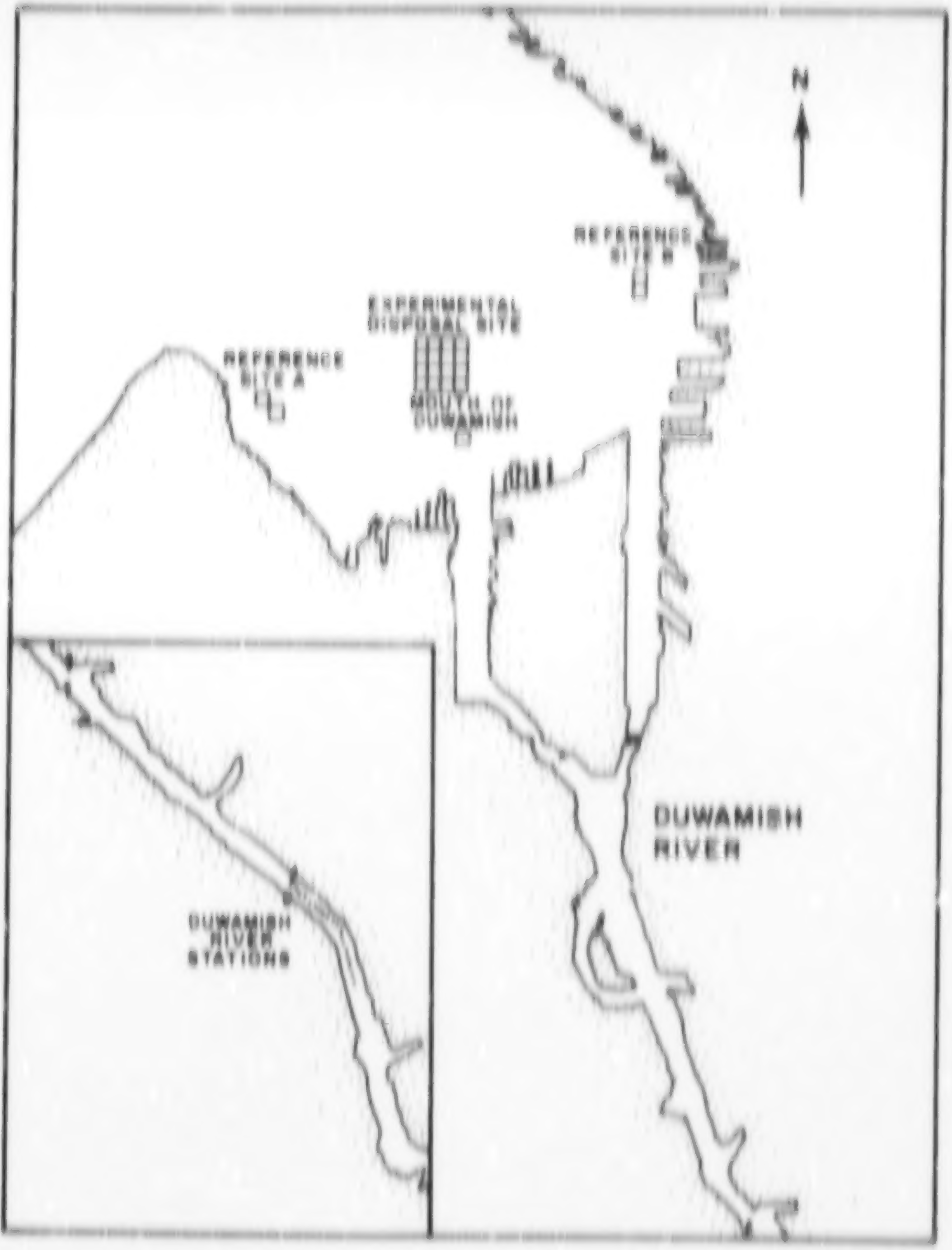

N
REFERENCE SITE B
EXPERIMENTAL DISPOSAL SITE
REFERENCE SITE A
MOUTH OF DUWAMISH
DUWAMISH RIVER
DUWAMISH RIVER STATIONS

Figure 2. Map of study area showing sampling sites

TABLE 3. SEDIMENT ANALYSES OF RIVER, SITE AND REFERENCE STATIONS BEFORE DISPOSAL OPERATIONS

Bulk Analyses (mg/kg dry wt.)

	River			Disposal Site			Reference		
	max	min	aver	max	min	aver	max	min	aver
Cd	<2	<2	<2	<2	<2	<2	<2	<2	<2
Cr	109	39	51	69	12	43	54	54	54
As*	70	35	51	43	19	28	33	23	28
Hg	0.7	0.1	0.3	0.6	0.2	0.4	0.6	0.2	0.4
Pb	120	<2	20	840	<12	105	500	<2	87

Interstitial Analyses

Metals (µg/l)	River			Disposal Site			Reference		
	max	min	aver	max	min	aver	max	min	aver
Cr	17	1	5.6	28	6	14.1	11	2	6.2
Hg	13.3	<1	6.5	8	<1	4.2	9	<1	4.5
Nutrients (mg/l)									
NH_3-N	40	0.6	19.3	4.0	0.4	2.1	2.5	0.8	1.6
Ortho P	1.69	<0.05	0.24	0.64	0.10	0.26	1.62	0.14	0.54

*neutron activation analyses

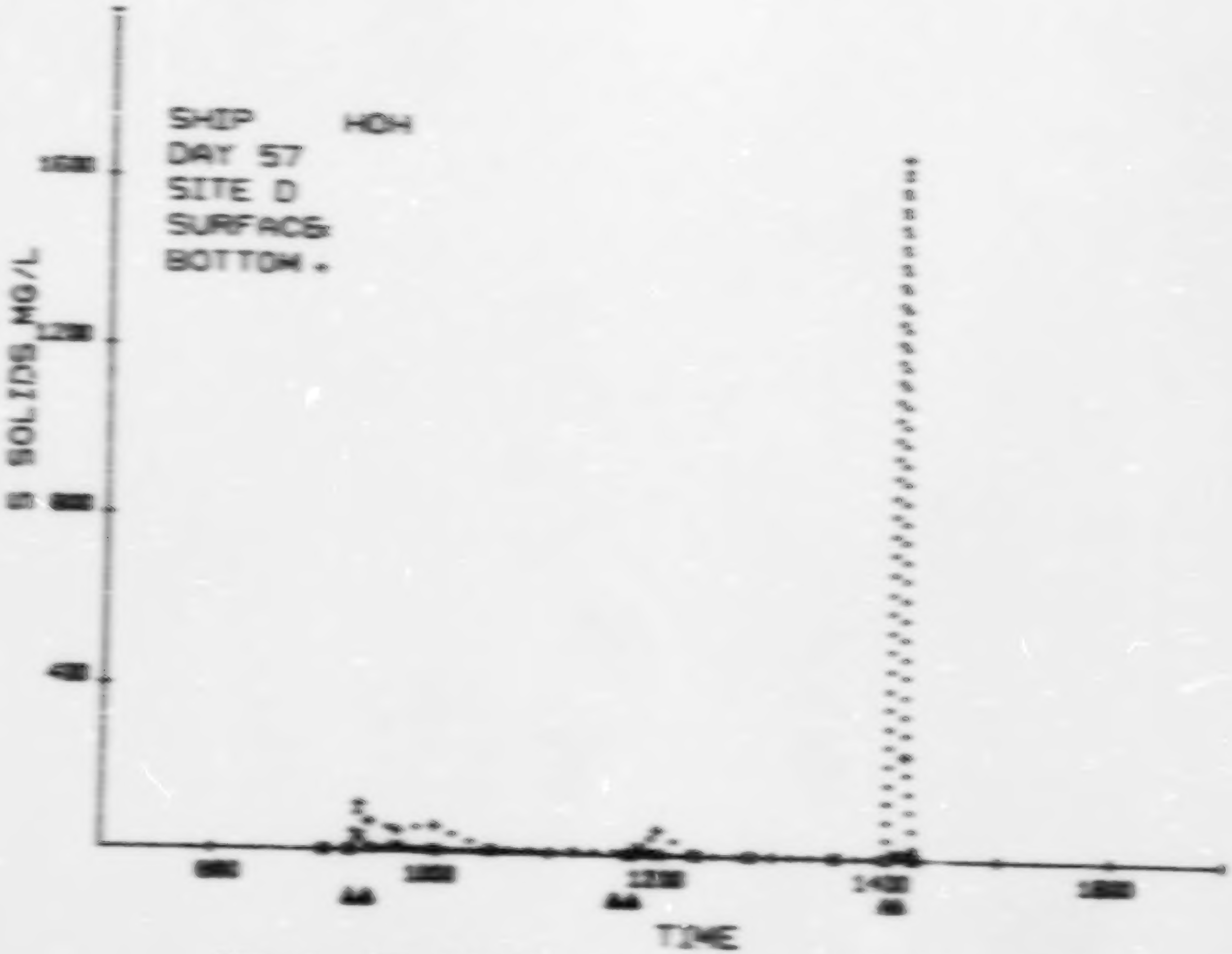


Figure 1. Suspended solids concentration in surface and bottom water during dumping.

material to the bottom rather than into the water column. There were, however, some short term effects noted in the water column. For example Figure 4 shows ammonia concentrations 1 m below the water surface and 1 m above the sediments in the immediate area of the disposal site during the disposal operation. A brief increase, especially in the bottom water was noted a few minutes after the dredged material was dumped. Ortho P concentration in the bottom water increased (Figure 5) immediately after the disposal, but in the surface waters there was a slight decrease. Minor fluctuations were noted in the NO_2-NO_3 concentration in the water column (Figure 6). An elutriate test in which one volume of wet sediment was shaken with four volumes of site water for 30 minutes under aerobic conditions, was performed on the river sediments. The results in Table 4 show that NH_3-N greatly increased in the site water after mixing with river sediment and NO_3-NO_2 showed only a slight increase. Ortho P, however, was removed from the site water by the sediment. Except for Ortho P the elutriate results agreed with the field observations.

TABLE 4. RESULTS OF ELUTRIATE TEST RUN ON RIVER SAMPLES

	Conc. in Site Water	Conc. in Interstitial H_2O	Conc. in Elutriate
Cr (µg/l)	4	7.4	3.5
Hg (µg/l)	<0.5	6.2	<0.5
NH_3-N (mg/l)	0.067	16.3	11.0
NO_3-NO_2 (mg/l)	0.426	---	0.49
Ortho P (mg/l)	0.076	0.24	0.014

Initial data on mercury concentration in the water column of Elliott Bay indicate that mercury was below the detectable level (0.5 µg/l). Some soluble chromium was noted in the water column during disposal as shown in Figure 7.

Algal assays (U. S. Environmental Protection Agency, 1974) using site water (control) and elutriate water (test) indicated that the net effect of sediment dumping on algal growth in Elliott Bay water results from the stripping of phosphorus from the water column, thus making the water, normally N or N and Si limited, phosphorus limited. The elutriate sample and elutriate plus 1.0 mg/l EDTA produced about the same amount of growth, both about 7% of the control (control = 34.6 mg/l algal dry weight, elutriate = 2.4 mg/l), indicating that heavy metals released from the sediment were probably not significant to algal growth.

Table 5 lists the concentrations of NH_3-N, Ortho P ang Hg found in the sediment interstitial water at the disposal site and reference stations before and after the disposal operation. Ammonia-N, which was an order of magnitude

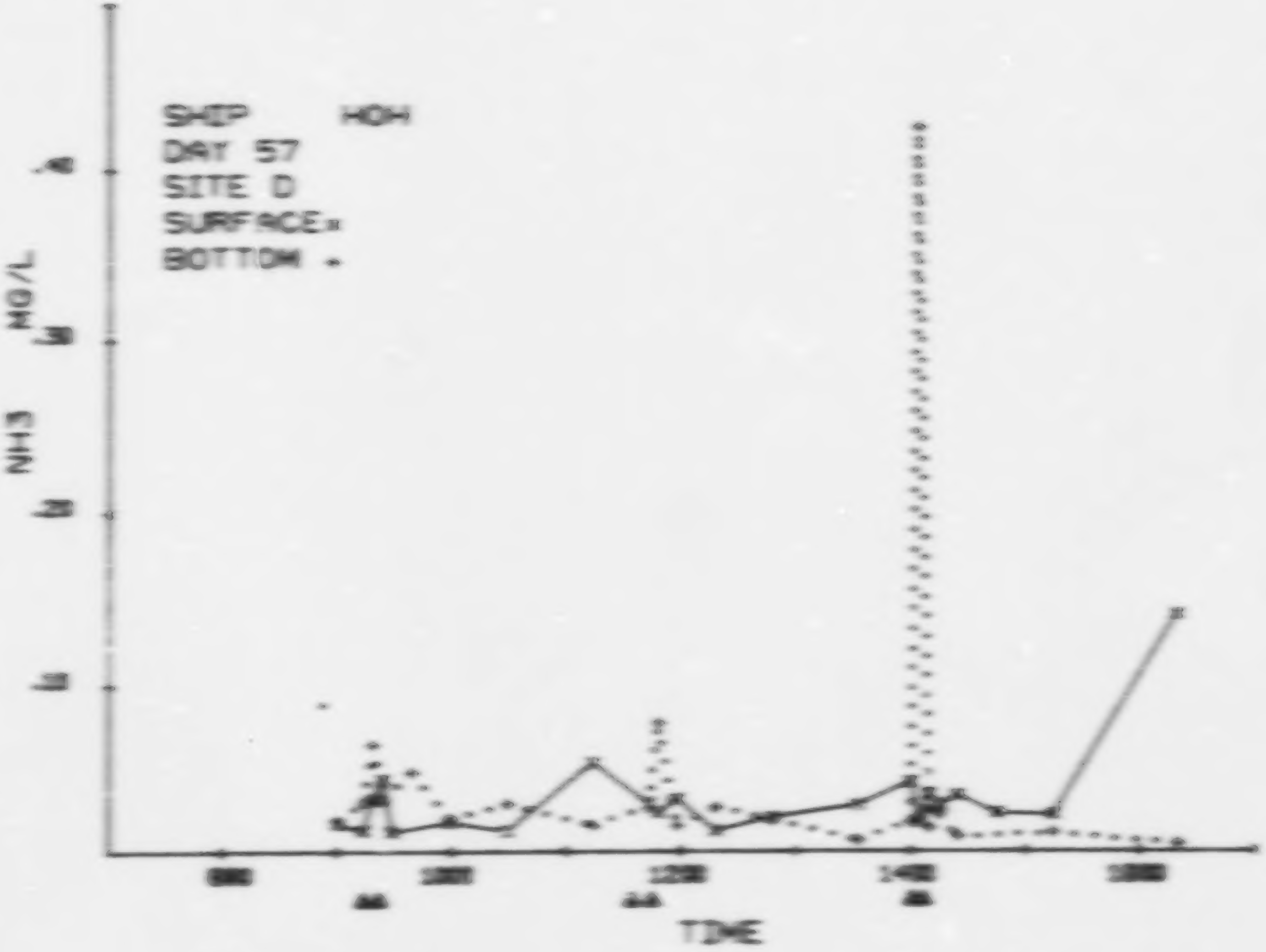


Figure 4. Ammonia concentration in surface and bottom water during dumping.

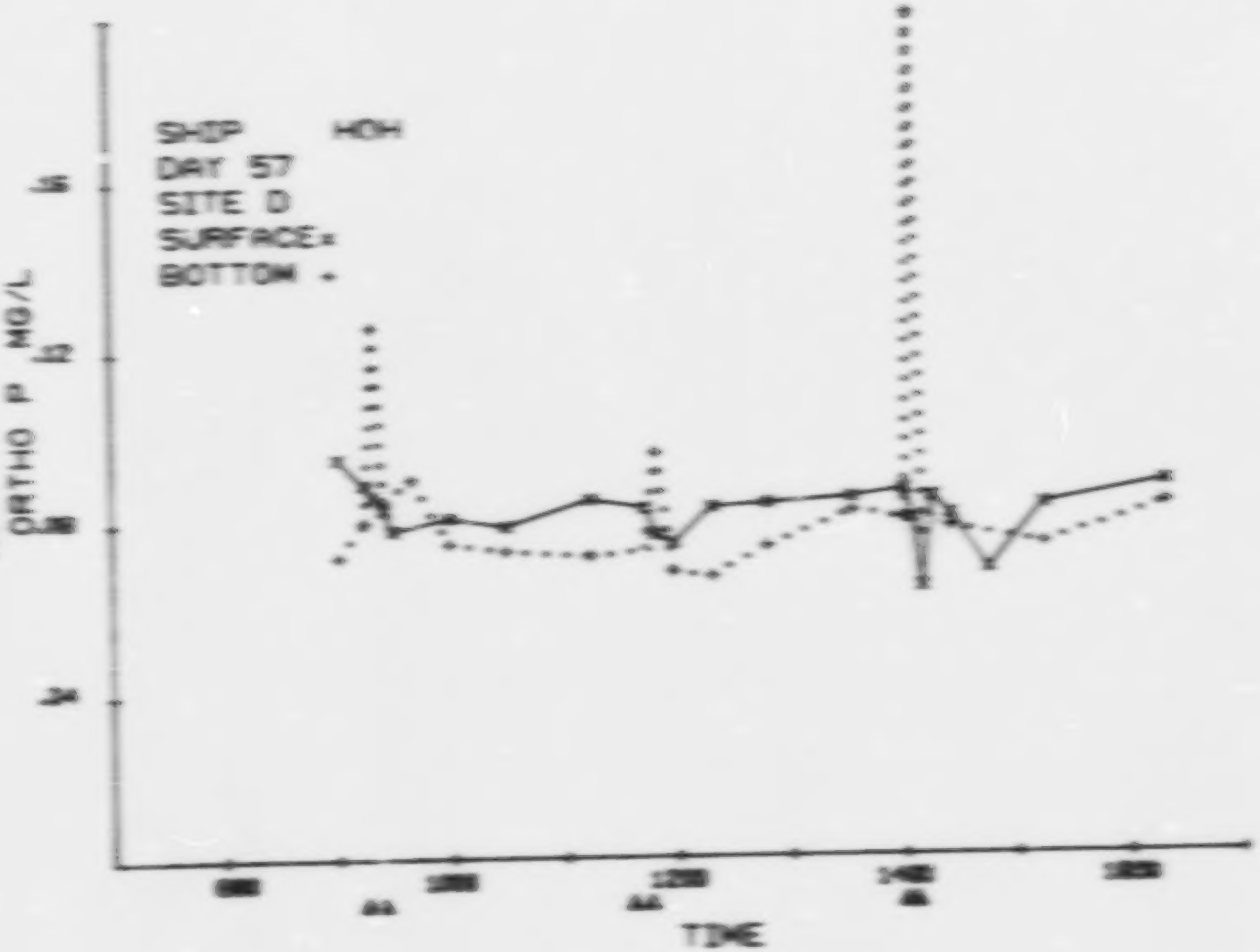


Figure 5. Ortho P concentration in surface and bottom water during dumping.

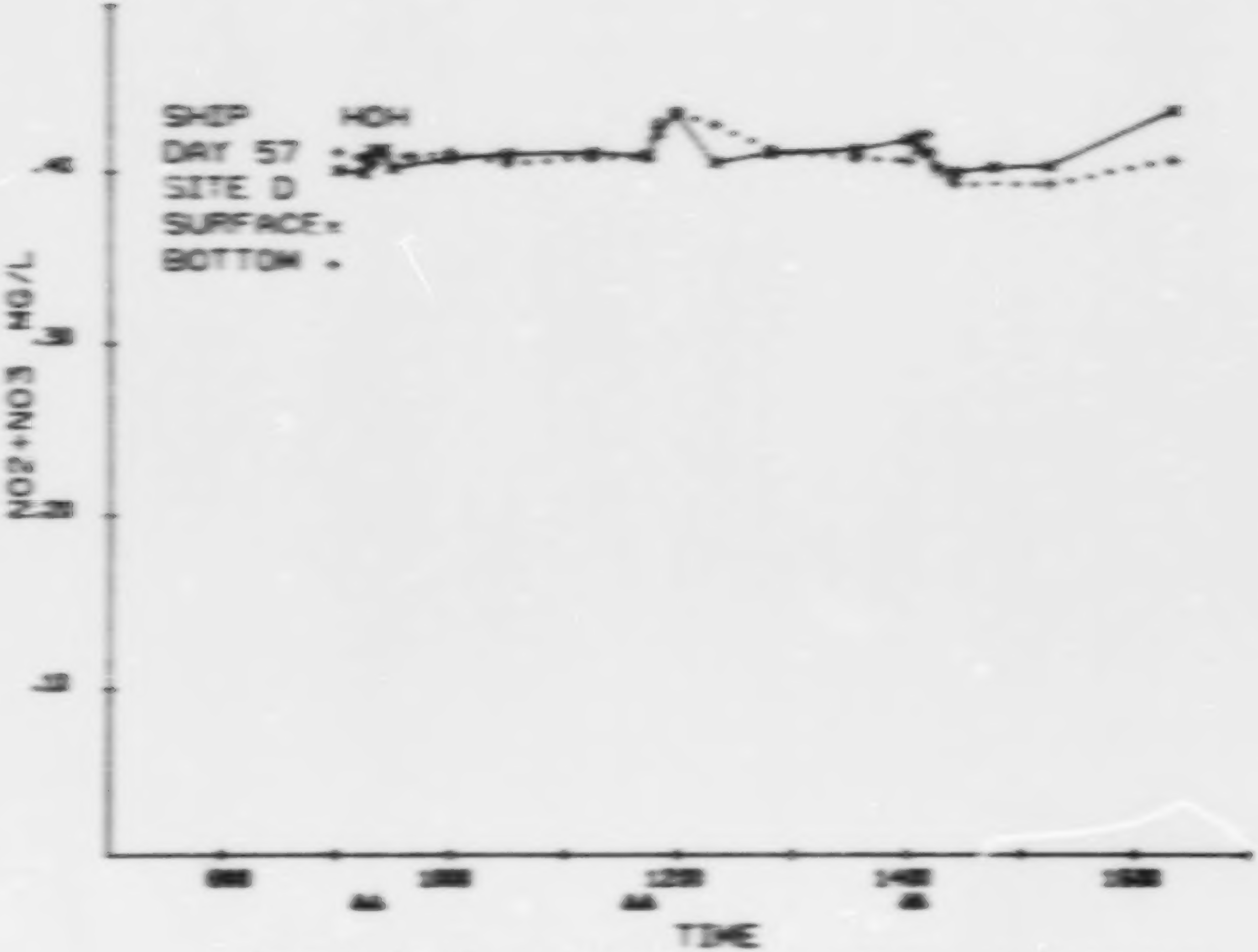


Figure 6. NO_2-NO_3 concentration in surface and bottom water during dumping.

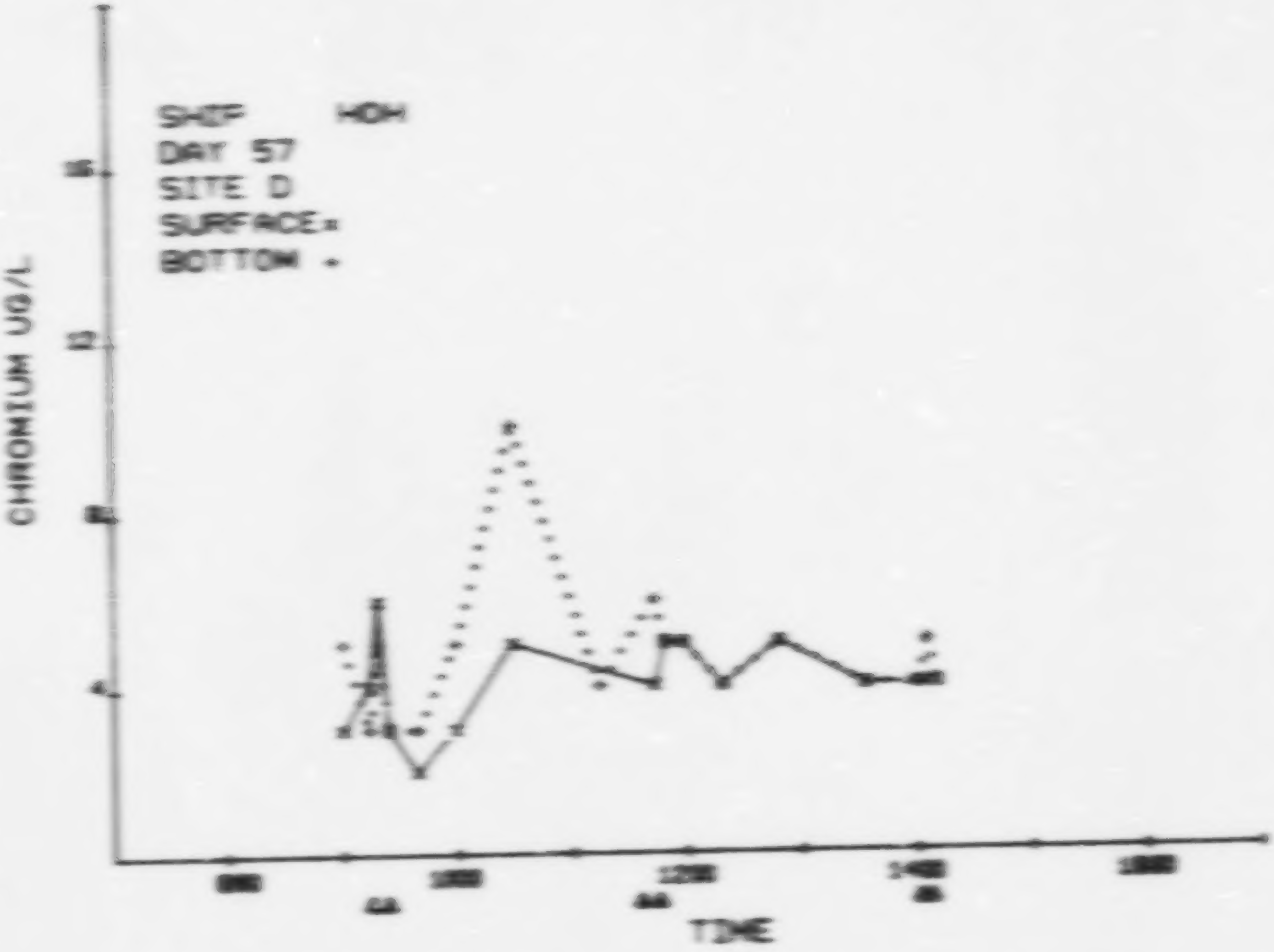


Figure 7. Soluble chromium concentration in surface and bottom water during dumping.

greater in the disposal material than the background bay sediments, increased greatly in the disposal site interstitial water after the disposal operation. One week after disposal higher concentrations were observed in the top 10 cm of the sediment when compared to the sediment below 10 cm; later the sediment below 10 cm had the higher concentration.

Mercury, likewise, was greater in the interstitial water immediately after the disposal operation but was near background levels after one month. The influence of the disposal operation on Ortho P in the interstitial water was not readily apparent.

TABLE 5. CONCENTRATION[a] OF CHEMICAL SPECIES IN SEDIMENT INTERSTITIAL WATER (IW) BEFORE AND AFTER DISPOSAL

Period	Disposal Site						Reference Sites		
	IW in top 10 cm			IW below 10 cm			IW in top 10 cm		
	NH_3	Ortho P	Hg	NH_3	Ortho P	Hg	NH_3	Ortho P	Hg
Before disposal	2.16	0.26	4.2	--	--	--	1.64	0.54	4.5
1 week after disposal	9.46	0.27	6.8	7.07	0.18	6.5	3.19	0.14	0.8
1 month after disposal	9.37	0.31	2.1	9.54	0.23	4.4	0.42	0.17	2.4
3 months after disposal	7.08	0.08	2.2	8.51	0.15	2.9	1.25	0.14	1.6

[a] NH_3 and Ortho P in mg/l

Hg in µg/l

ACKNOWLEDGEMENTS

The Duwamish Waterways project was designed and funded by the U. S. Army Corps of Engineers Environmental Effects Laboratory of the Waterways Experiment Station, Vicksburg, Mississippi. Drs. Jeff Johnson and Robert Engler of the Experiment Station were responsible for initiating and coordinating the research. The authors would also like to thank R. Manabe, K. Harris, and M. Tollefson for analyzing the samples that are used in this report. We also wish to thank C. Powers, B. Lauer, D. Taug, and J. Carkin for their efforts in collecting and preparing samples and processing data.

REFERENCES

U. S. Environmental Protection Agency. 1973. Water Quality Criteria 1972. EPA-R3-73-033 594 p.

U. S. Environmental Protection Agency. 1974. Marine Algal Assay Procedure: Bottle Test. National Environmental Research Center, Corvallis, Oregon EPA-660/3-75-008 43 p.

DREDGING CONDITIONS INFLUENCING THE UPTAKE OF HEAVY METALS BY ORGANISMS

J. F. Sustar and T. H. Wakeman
U.S. Army Engineer District, San Francisco
211 Main Street
San Francisco, California 94105

ABSTRACT

Studies were conducted by the San Francisco District of the U.S. Army Corps of Engineers during the period 1971 through 1976 to evaluate the impacts associated with dredging and sediment release at open water disposal sites in San Francisco Bay. Although significant changes were observed in dissolved oxygen reductions, suspended solids increases, and trace elements, chlorinated hydrocarbon and nitrogen (nitrate and ammonia) releases, the changes were not found to be synonymous with biological impacts. Uptake and desorption of trace elements by organisms were observed. Contaminant levels in estuarine organisms appear to be controlled by a limited number of factors. Suggested factors are the long-term process of sediment resuspension-recirculation, seasonal fluctuations in salinity and sources of contaminants both man introduced and geologic formation.

INTRODUCTION

During the late 1960's, an awareness of man's impact on the environment and, in turn, on man himself became a focal point not only for new projects but also for the maintenance of existing projects. Within an estuarine system, the development of facilities for trade along with associated services is the most noticeable impact. The continuance of the development is contingent on the availability of deep water. This means dredging. The dredging and disposal operation can be viewed from two different aspects. First, the dredging and disposal operation is a separate and distinct activity. Second, the dredging and disposal operation can be viewed in terms of associated changes in the system, that is, the resulting channel. Aside from the land development, this means changes in the estuarine regimes in terms of water circulation, salinity, sediment type, contaminant levels and contaminant sources.

From 1971 through 1976, the San Francisco District of the U.S. Army Corps of Engineers conducted studies to assess the impacts associated with maintenance dredging in San Francisco Bay with open water disposal of the sediments. The study addressed the first view of the dredging and disposal operation and attempted to advance the understanding of the interrelationships between the physical, chemical and biological systems in the Bay. The various systems were initially studied independently to form a characterization of the Bay. These studies were followed by investigations of how the extreme ranges of the various systems influence other systems, primarily, biological.

DESCRIPTION OF THE BAY

San Francisco Bay is a drowned valley through which passes the drainage of the great Central Basin of California. The outlet to the Pacific Ocean is the Golden Gate, 1.6 kilometer wide, 4.8 kilometer long strait with depths in excess of 90 meters. The Bay system is composed of several distinct areas separated by narrow straits. Suisun Bay at the upper end is moderately narrow and allows runoff from the Central Valley to pass quickly into the more saline areas west of the 11 kilometer long Carquinez Strait. San Pablo Bay provides the first area of extensive mixing of freshwater runoff with saline ocean water. The isolated South San Francisco Bay receives very little runoff due to no large tributaries and several impoundments of small local drainage area. A pronounced wet and dry season is characteristic of the area with about 85 percent of the total rainfall occurring between November and April. The Bay system has an area of 1,026 square kilometers at mean lower low water and 1,191 square kilometers at mean higher high water, leaving extensive mudflats exposed at lower low water. The Bay is generally shallow with two-thirds of the area less than 5.5 meters deep and only 20 percent greater than 9 meters deep.

DREDGED SEDIMENT

With the exception of the sandy sediments associated with the San Francisco Bay Channel, Southhampton Shoal, Pinole Shoal and Suisun Bay, maintenance dredging operations in the Bay move "Younger Bay Mud." Bay mud consists of soft, plastic, black-to-gray silty clay or clayey silt with minor organic material and clayey fine-grained sand which has been deposited in the Bay largely due to flocculation. A typical grain size is as follows:

	Dispersed	Non-dispersed
% Sand (>0.075 mm)	12	13
% Silt	46	87
% Clay (<0.002 mm)	42	0

Organic carbon content is about one and one-half percent and *in situ* density is about 1.3 to 1.4 grams per cubic centimeter. The clay size fraction is composed of one-third montmorillinite, one-third normal and hydrated mica, and one-third mixed-layered montmorillinite, chloritic and kaolinitic materials.

Sediment deposition patterns reflect the energy gradient formed by dynamic estuarine forces within the Bay. Deposition zones are situated in low energy areas where the energy of wave action and current velocity is dissipated or nonexistant.

Contaminants enter the San Francisco Bay system through natural weathering processes of rocks and soils and by anthropogenic means on land, air and water. Dissolved substances are sorbed by particulate matter both before entry and after entry into the estuary. These organic and inorganic contaminants show behavior and distribution patterns similar to that of natural sediments with the physical setting and estuarine processes responsible for their movement and deposition. Mean concentrations of contaminants in the Bay based on Corps of Engineers sampling are as follows:

	Dredged Channels	Undredged Areas
Lead ppm	35.5	34.3
Zinc ppm	108.1	110.1
Mercury ppm	0.55	0.71
Cadmium ppm	1.59	0.86
Copper ppm	41.6	36.2
Oil and grease ppm	800	500
Volatile solids ppm x 10^4	6.03	5.65
COD ppm x 10^4	1000	1000

Contaminant levels are generally associated with sediment type (particle size) which is reflected in both vertical and horizontal distribution of contaminants. This relationship is not absolute and other factors such as proximity to source of contaminants, rate of shoaling, rate of contaminant input and association to other parameters such as organics play a role in the distribution. Highest contaminant levels are normally associated with the finest sediments. As such, the higher concentrations of contaminants in dredged channels can be attributed to the finer grain size associated with maintenance dredging. Since dredged channels are out of equilibrium, forming a lower energy regime, finer sediments will tend to shoal.

About 7.6 million cubic meters of sediments are dredged annually in San Francisco Bay. The majority of the dredging is accomplished with a trailing suction hopper dredge with maintenance of berthing facilities and marinas by clamshell and hydraulic cutterhead dredges.

SEDIMENT RESUSPENSION AND INTERACTION WITH WATER

The type of sediment and the degree to which it is disturbed determine the sediment resuspension during dredging and the immediate release pattern during disposal at open water sites. The disturbance including the adding and mixing with water depends on the type and size of dredge, the efficiency of operation and the configuration of the shoal. With both hopper dredge and clamshell dredge, the plume can extend more than 700 meters downstream. Concentrations at overflow ports were measured as high as 8.7 grams per liter. These concentrations are reduced quickly to the hundred milligram per liter range. At the sediment-water interface, higher concentrations in the range of 2 grams per liter were measured.

The disturbance during open water release is limited to the bottom few meters of the water column regardless of whether the sediment mounds or disperses. Complete mounding of cohesive, new construction sediments excavated by clamshell was observed at a 100-fathom disposal site. Maintenance sediments excavated by hopper dredge were found to leave an in-bay disposal site typically within fifteen minutes of release, being quickly assimilated into the Bay sediment regime. In the case of mound, very little contact time and area occurs between the sediment and water column, limiting the immediate interaction of contaminants with the water column. With dispersal, contact time and area with the upper water column is similar to that of mounding. However, in the bottom two meters, sediment concentrations were immediately dispersed to a transport concentration of about ten grams per liter for single plume movement and about twenty grams per liter for interaction of a twin plume as with a hopper dredge with two hoppers. With in-bay disposal, the sediments were distributed over a 260 square kilometer study area to depths in excess of 23 centimeters. The majority of the samples in the study area had less than four percent dredged sediment.

Dredging and disposal in the Bay were not observed to cause significant changes in conductivity/salinity, temperature of pH. In addition to suspended solids increases, water quality changes which were observed to be significant included dissolved oxygen reductions, and trace element, chlorinated hydrocarbon and nitrogen (nitrate and ammonia) releases. During dredging dissolved oxygen reductions were found to occur only about 25 percent of the time. The reduction is about two parts per million and lasts for about two minutes before returning to background, typically eight to nine parts per million. Reductions at the sediment-water interface were as much as four parts per million for about eight minutes. The upper water column during open water disposal experienced reductions similar to the dredging operation. Near the bottom in the base surge cloud, significant oxygen depletion was observed. Reductions of up to six parts per million were observed. Ambient concentrations were regained after an average of three to four minutes, but could be influenced for as long as eleven minutes. With sediments having a lower oxygen consuming material, increases in oxygen were observed.

The result of a selective extraction scheme indicated that a significant portion of trace elements investigated are in the residual phase of the sediment. They were mercury (77 percent), iron (60 percent), manganese and zinc (54 percent), copper (53 percent), lead (50 percent) and cadmium (7 percent). Phases other than in the residual or lattice of the sediment are available for release under proper environmental conditions. The two factors which exert the greatest effects are pH and oxidation-reduction potential of the ambient environment. The pH of the Bay system is near neutrality and the dredging with open water disposal was not observed to cause a change. Of significantly greater importance is the effect of oxidation-reduction potential shifts. Significant amounts of metals were found in the organic and sulfide-like phases using a hydrogen peroxide treatment. They were cadmium (92 percent), lead (15 percent), copper (43 percent), zinc (39 percent), mercury (23 percent) and iron (19 percent). Bay sediments contain 1,000-3,000 parts per million of sulfides and thus represent a reducing environment with trace metals associated with sulfides expected. In sorption-desorption experiments, oxidation-reduction potential was found to have the greatest effect on trace metal fate. Under oxygen rich conditions, significantly more copper, cadmium, lead and zinc were found in the elutriate that under reduced conditions. Iron acted in an opposite manner. Higher salinity elutriates significantly influenced higher cadmium and zinc concentrations in oxygen rich samples and iron in oxygen deficient conditions. Agitation time significantly affected the release of cadmium, copper and zinc under oxidizing conditions. More metals were released at the longer shaking periods, suggesting that a kinetic mechanism may play a role in the fate of trace metals. The release of trace elements (cadmium, copper, lead and zinc) under oxygen rich conditions in general increased the water column concentration 30 to 200 percent. Ratios of elutriate to original water concentrations in general ranged from 1.3 to 2.0 for samples which originally had trace metal concentrations similar to values found in the bay.

In field monitoring, solute concentrations of cadmium, copper and lead increased in the release plume by 6, 4 and 9 times respectively. Cadmium concentrations increased from 0.22 parts per billion (ppb) to 1.35 ppb, copper from 1.29 ppb to 5.0 ppb and lead from 0.21 ppb to 1.88 ppb. These observed increases lasted less than one and one-half hours which was the frequency of sampling.

BIOLOGICAL UPTAKE

Chemical reactions which might be biologically significant during dredging and disposal operations are not as intuitively obvious as impacts due to physical distrubance. During both laboratory and field investigations, significant changes in water quality were demonstrated owing to chemical reactions occurring from Bay sediment resuspension. No analogous changes in organisms were observed. Water quality impacts were not found to be synonymous with biological impacts.

Field studies were conducted to determine uptake of silver, cadmium, copper, mercury, nickel, lead, selenium, zinc, and arsenic by invertebrates *Macoma balthica*, *Neanthes succinea*, *Ampelisca milleri* and *Ischadium demissum* and the mussel *Mytilus edulis*. Two periods of heaviest rainfall for the year causing pronounced decreases in salinity coincided with the two dredging periods. Metal concentrations in sediments and invertebrates fluctuated during the period of study. With the exception of nickle concentrations in *N. succinea*, no significant changes in metal levels were associated with dredging activities although significant uptake and desorption did occur throughout the study area. The changes in nickel were significantly greater at stations outside the dredging zone suggesting that dredging inhibited nickel accumulation in this species.

The results of associated nine day studies showed that the greatest uptake and accumulation in *M. balthica* of the chloride salts of silver, cadmium, copper, mercury and lead occurred with the highest concentrations of these metals in the lowest salinity water. The greatest desorption of nickel, selenium zinc and arsenic occurred in the clams exposed with the highest salinity.

A second study was conducted during a disposal operation. The concentration of twelve trace elements (Ag, As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Se, and Zn) were monitored in sediments, suspended and settled particulates, selected benthic invertebrates (*Macoma nasuta*, *Pectinaria californiensis*, *Stylatula elongata*, *Tritonia diomedia*, *Glycera americana*, and *G. robusta*) and transplanted mussels, *Mytilus edulis*. The experimental disposal operation did not affect trace element concentrations in the benthic invertebrates nor in mussels transplanted to the disposal area.

Bay dredging and disposal activities were found to redistribute contaminated sediments without, under the conditions of the experiments, resulting in increased contaminant availability. Fluctuations in the concentrations of test elements were highly correlated with each other in sediment, invertebrates, suspended and settled particulates, although element levels within any one component were not correlated with element levels in any other component. The high correlation among trace elements within each component suggests that only one or a few parameters may control trace element fluxes in San Francisco Bay.

CONCLUSIONS

Based on results of studies conducted in San Francisco Bay during the Dredge Disposal Study, the following conclusions have been formulated regarding trace element uptake by organisms.

Although significant changes in water quality during dredging with open water disposal were demonstrated, no analogous changes in organisms were observed. Thus, water quality impacts were not found to be synonymous with biological impacts.

Release of trace elements during dredging and disposal operations seem to be of sufficiently low leveles and last for such short durations that their availability for uptake and accumulation is extremely limited.

Salinity increases significantly intensify the potential for release of certain trace elements from resuspended sediments. Organisms have been observed to have greater uptake rates during periods of decreased salinity and greateer depuration rates in high salinity water. These two opposing conditions suggets that there is potentially a natural defense mechanism operating in organisms to safeguard them from excessive trace element accumulation.

Contaminant levels in estuarine organisms appears to be controlled by a limited number of factors. Suggested factors are the long-term process of sediment resuspension-recirculation, seasonal fluctuations in salinity and sources of contaminants both anthropogenic and geologic. The biological impact may be dependent on the form of contaminant and whether or not the sediment system can assimilate the contaminant loading. With the observed sorption-desorption by organisms and the fluctuating conditions in the estuary, impacts such as high accumulations, mutations and toxicity would not be expected unless the contaminant loading is foreign, in the case of synthetic chemicals, or above the assimilation capability of the estuary with the associated sediment regime, in the case of a low energy regime in which the changes in ambient conditions is great.

ACKNOWLEDGEMENT

This paper is based on the Dredge Disposal Study - San Francisco Bay and Estuary by the U.S. Army Engineer District, San Francisco. Publication of this paper has been approved by the Corps of Engineers but the expressed views, interpretations and conclusions are those of the authors and do not necessarily represent those of the U.S. Army Corps of Engineers. Added thanks is given to Mr. Richard Ecker, a memember of the District task force and to the Corps personnel and many contractors who conducted the individual study elements.

DREDGED MATERIAL DENSIFICATION AND TREATMENT OF CONTAMINATED DREDGED MATERIAL

C. C. Calhoun, Jr.
Manager, Disposal Operations Project
Environmental Effects Laboratory
USAE Waterways Experiment Station
P. O. Box 631
Vicksburg, Mississippi 39180

ABSTRACT

This paper describes work conducted by the Disposal Operations Project of the U. S. Corps of Engineers' Dredged Material Research Program to develop methods for dredged material densification and treatment of contaminated dredged material. Densification techniques include conventional gravity drainage, mechanical agitation, electro-osmosis, crust management, aeration, frost action, wicks, and vegetation. With the exception of aeration and frost action, all techniques are being evaluated by large-scale field tests. The applicability of various conventional wastewater treatment methods to dredged material is discussed. The extent of oil and grease contamination associated with confined disposal areas is reviewed. The use of flocculants to improve the quality of effluents for confined containment areas is outlined. Field tests to determine the effectiveness of injecting oxygen and air into the pipeline during open-water disposal to increase the dissolved oxygen in the water column are discussed as well as field tests to evaluate the ability of vegetation to remove contaminants from disposal area effluents.

INTRODUCTION

The Corps of Engineers' Dredged Material Research Program (DMRP) is a comprehensive five-year research program initiated in March 1973 to address problems associated with the disposal of dredged material. The program is being administered by the Environmental Effects Laboratory (EEL) of the Waterways Experiment Station (WES), Vicksburg, Mississippi. The DMRP is divided into four projects. This paper describes work being conducted within the Disposal Operations Project to densify dredged material and to treat contaminated dredged material. The densification studies are under the direction of Dr. T. A. Haliburton while the treatment studies are being directed by Mr. T. K. Moore.

DREDGED MATERIAL DENSIFICATION

Background.

In most cases where fine-grained dredged material from harbors and waterways is placed in slurry form into containment areas, the material remains at high water contents for years. The large volume of trapped water significantly reduces the capacity of the containment area to retain solids. If significant quantities of this water can be removed, the service life of containment areas will be extended and the problems associated with securing new disposal areas reduced. Therefore the purpose of densification is to gain additional volume and not to stabilize the area per se.

The feasibility, both economic and technical, of dewatering or densifying dredged material is being addressed by DMRP Task 5A (Dredged Material Densification). The major thrust of the work in this task was initiated in October 1974 by a planning symposium attended by a panel of experts in various fields dealing with the removal or movement of water in soils and sludges as well as dredging experts. From the symposium, guidance was given for establishing a program for evaluating in the laboratory or by small-scale field studies the feasibility of various techniques for dewatering dredged material. Although there are many techniques for dewatering dredged material, the feasibility of these methods is usually controlled by the economics of dewatering vast areas. The dewatering of a single containment area may involve hundreds to perhaps thousands of hectares. Consequently, the costs of applying to containment areas conventional techniques used to economically dewater building foundations or sludge fields may be prohibitively high. Consideration must also be given to the characteristics of the site itself when selecting a dewatering technique. Techniques that may be applicable to active sites where the depth of the material is not great may not be applicable to sites nearing capacity or previously abandoned when filled with material 9 to 12 m deep. All of these factors were considered when decisions were made concerning the techniques to be evaluated.

A second symposium was held approximately one year later and the panel evaluated the results of the laboratory and small-scale field studies and made recommendations as to which techniques deserved further evaluation in the field. The various studies (work units) considered at the second symposium are shown in Table 1. In addition, one work unit conducted prior to the first symposium and a work unit in a related task are listed and discussed.

TABLE 1: DREDGED MATERIAL DENSIFICATION PRELIMINARY AND LABORATORY INVESTIGATIONS

Work Units	Contractor*
4A16 Performance of Containment Areas Filled with Dredged Material	Massachusetts Institute of Technology; Cambridge, Massachusetts
5A01 Methodology for Dredged Material Reclamation and Drainage	Dames & Moore, San Francisco, California
5A02 A Laboratory Study of Dredged Material Slurry Water Loss Due to Mechanical Agitation	EEL, MESL
5A03 State-of-the-Art Survey and Evaluation of Current Physical, Mechanical, and Chemical Dewatering and Densification Techniques	SPL
5A04 A Laboratory Study to Determine the Variables that Influence the Electro-Osmotic Dewatering of Dredged Material	KMA Research Institute, Mesa, Arizona
5A05 A Laboratory Study of Aeration as a Feasible Technique for Dewatering Fine-Grained Dredged Material	Environmental Engineering Consultants, Inc., Stillwater, Oklahoma
5A06 Feasibility Study of General Crust Management as a Technique for Increasing Capacities of Dredged Material Containment Areas	Texas A&M University, College Station, Texas
5A07 Feasibility of Frost Action for Densification of Dredged Material	USACE Cold Regions Research and Engineering Laboratory; Hanover, New Hampshire
5A10 Development of Capillary Enhancement Devices for Dewatering Fine-Grained Dredged Material	SPL

*WES - Waterways Experiment Station
EEL - Environmental Effects Laboratory (WES)
MESL - Mobility and Environmental Systems Laboratory (WES)
SPL - Soils and Pavement Laboratory (WES)

Preliminary and Laboratory Investigations

4A16A

In order to estimate the effectiveness of a densification technique, the volume the material will occupy if it is placed in the containment area and nothing is done to densify it must be known. In this work unit, as a part of another task, a methodology is being developed for determining the capacity of containment areas when conventional filling and operations are used. This methodology will provide the "do-nothing" case and be the datum from which the effectiveness of the densification techniques can be determined.

5A01

The initial study within Task 5A was conducted in 1973 prior to the first planning symposium and out of phase with the other studies since it was a field demonstration of a technique previously used to densify dredged material. Basically, the technique was to agitate the material continuously to accelerate the loss of water by evaporation. In the early stages of the DMRP, it was noted that practically all fine-grained material in containment areas exhibited a relatively thin crust of dried, firm material underlain by soft, very wet material. It was assumed that the formation of the crust effectively sealed underlying material from the evaporation process and, therefore, that agitating the material would prevent the crust from forming and allow evaporation to continue. Although there appeared to be a significant increase in the rate of dewatering of the agitated material when compared to material not agitated, the field conditions did not allow rigorous analysis of the data. Consequently, smaller scale controlled tests (5A02) were performed and followed up by a relatively large-scale test.

5A02

The results of the controlled tests indicated that there was no significant difference in the rate of water removal from agitated and non-agitated fine-grained material. In the areas where the material was not agitated, desiccation cracks formed and extended to the bottom of the material. It appeared that the formation of the cracks allowed the evaporation process to continue at about the same rate as from the agitated material.

These findings still do not answer why, under the field conditions observed previously, a crust formed and underlying material remained wet for long periods of time. Upon closer investigation of various sites where the crust formed, it was observed that the containment area acted as a "bathtub" and most water in the site could not be removed except by evaporation. It became apparent that a water table was maintained by recharge from rain or other sources and that the desiccation cracks in the crust extended only to the elevation of this perched water table. This was also reported in a review of tests conducted by private industry on a fine-grained industrial waste (5A03). Results from Work Unit 5A06 also tend to verify the influence of the perched water table on the formation of the crust. Based on the information developed from this study, the DMRP no longer recommends continual agitation to accelerate the dewatering of fine-grained dredged material.

5A03

In this work unit, a review was made of the performance of conventional dewatering techniques used in soil mechanics and foundation engineering and techniques used to dewater industrial sludges and slimes. Results from this investigation indicate that the use of various types of subdrains fed by gravity drainage or accelerated drainage from applied vacuum is feasible. This study also revealed that dewatering low plasticity material (silt) yielded little additional volume, but significant volume could be gained by dewatering more plastic material (clays).

5A04

Electro-osmosis is most often used for soil stabilization where small amounts of water must be removed in a short period of time. Consequently, large voltage gradients on the order of 1 V/cm are used for relatively short periods of time (weeks). To dewater dredged material, relatively large volumes of water must be removed, but normally relatively long periods of time are available (months to years). This study considered the effects of using electro-osmosis at low voltages applied continuously or intermittently for long periods of time. The results of the study indicated that the dredged material could be dewatered at gradients as low as [illegible] V/cm and that the process continued if the voltage was applied intermittently. These findings are significant since power costs are drastically reduced, thus resulting in the overall cost of the process being much lower than the cost of the conventional electro-osmosis process. Also, the cost could be further reduced by applying the current intermittently during off peak periods or by generating electricity from sources such as windmills.

5A05

Although conceptually the forcing of air up from the bottom of the dredged material would greatly accelerate water removal, the process has limited feasibility. It is anticipated that the process would be used where periods between fillings are short and where, because of limited space, water must be removed in a very short period of time.

5A06

An in-depth study of crust development is presently underway. Since the crust provided by nature entails no expense and results in a very dense material, the processes involved in its formation are being investigated and methods of accelerating crust formation through management of the area are being developed. A methodology will be provided to predict the rate of crust formation.

5A07

It is a well-known phenomenon that the permeability of a soil is greatly increased from that of its natural state by freezing and then allowing it to thaw. In many parts of the world, dredged material is frozen to a depth of several feet during winter. If water from the material could be removed during the spring thaw, significant consolidation would occur. In this work unit the magnitude of this consolidation is being investigated and methods of removing the water developed.

Probably the most unconventional dewatering technique being considered is the use of capillary enhancement devices or wicks. Wicks provide a path for water within the dredged material to rise to the surface where it evaporates. Preliminary results indicate that the rate of dewatering is accelerated considerably by wicks.

Field Studies

In 1975 the DMRP entered into a cooperative agreement with the Mobile District of the Corps of Engineers for a full-scale field demonstration of various densification techniques at the Upper Polecat Bay (UPB) disposal area. The disposal area is located along the Mobile River in Mobile, Alabama, and is used to contain material dredged from the main channel and harbor. The disposal area is approximately 34 hectares in size and contained 2.5 to 3 m of dredged material in 1975. The site was created in 1970 and dredged material was placed in the area in 1970 and 1972. The material was predominantly an organic clay sediment of high plasticity. The foundation material was very soft. It is anticipated that the increased effective load resulting from dewatering the overlying material will consolidate the foundation material significantly. The consolidation will also result in additional volume available to contain dredged material. A layout of the demonstration area is shown in Figure 1.

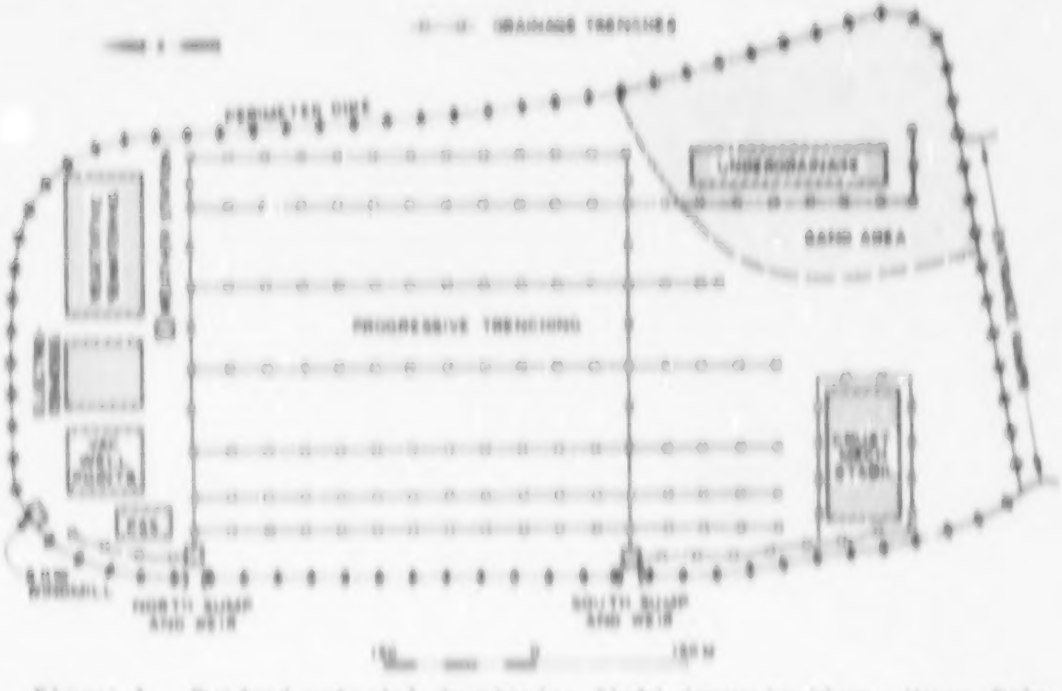


Figure 1. Dredged material dewatering field demonstrations, Upper Polecat Bay Disposal Area, U. S. Army Engineer District, Mobile

When dewatering operations were initiated by the DMRP in August 1975, the site conditions in all but the southeast corner consisted of a 5- to 15-cm crust over the soft dredged material. This underlying material had the consistency of warm axle grease and water contents of 1 to 2 times its liquid limit. About 80 percent of the site was under ponded surface water. Little vegetation existed.

Ponded water was removed from the site using the Riverine Utility Craft or RUC (Photo 1). The RUC, a 5440-kg, 6.1-m-long twin helical screw amphibious vehicle, has superior mobility in soil of extremely poor support conditions. It was originally developed for military purposes to fill mobility gaps between boats and conventional tracked or rubber-tired vehicles.

Photo 1. RUC operating in disposal area

The RUC creates two semicircular ruts or trenches as it moves across soft ground, thereby providing effective drainage channels. Approximately 46 cm of ponded water was removed from the UPB site and a nearby 97-hectare site in less than two days using the RUC to trench, cut through high spots, and clean areas around weirs. It was estimated that the equivalent of several thousand dollars worth of dragline work was performed under conditions where dragline operation would have been difficult if not impossible.

As shown in Table 2, a total of seven Task 5A field demonstration work units as well as one field study in a related task are presently scheduled to be carried out at the UPB disposal area. The location of some of the demonstrations are shown in Figure 1. Each of the techniques will be discussed and when possible, costs will be estimated.

TABLE 2. DREDGED MATERIAL DENSIFICATION FIELD DEMONSTRATIONS

Work Units	Contractor*
5A08 Densification of Dredged Material by Progressive Trenching	EEL
5A09 Consolidation of Fine-Grained Dredged Material with Windmill-Powered Well Points	Mobile District, EEL
5A11 Injection of Sand Slurry into Fine-Grained Dredged Material	SPL; EEL
5A14 Mechanical Stabilization of Dredged Material	EEL; MESL
5A15 Dewatering by Underdrainage	SPL
5A16 Electro-Osmotic Dewatering Field Study	Mobile District (L. Casagrande and E. E. O'Bannon, Consultants)
5A18 Vegetation Dewatering	Dauphin Island Sea Laboratory
2C09 Field Evaluation of Equipment for Operation in Containment Areas	MESL

*WES - Waterways Experiment Station
EEL - Environmental Effects Laboratory (WES)
MESL - Mobility and Environmental Systems Laboratory (WES)
SPL - Soils and Pavement Laboratory (WES)

5A08

This work unit being conducted over the center 24 hectares of the site is concerned with determining the effect of progressive trenching on the drying and drainage rate of fine-grained dredged material. The beneficial effects will result from rapidly removing precipitation, lowering the perched water table, and promoting desiccation and surface crust formation. Equipment used in the trenching study has included the RUC (with and without implements), an amphibious dragline, and a conventional dragline on mats. Starting from the conditions described earlier, the progressive trenching so far has resulted in the development of a surface crust from 0.5- to 1.5-cm thick.

5A09

A 6-kw windmill with a three-bladed 5-m-diameter prop located on a 12-m tower is being used to provide electricity to run vacuum and water pumps in an attempt to dewater and consolidate fine-grained dredged material with vacuum well points. It is believed that dewatering constraints inherent in the relatively low hydraulic permeability of dredged material slurry may be somewhat nullified by relatively long-term dewatering, as opposed to more conventional short-term dewatering uses of vacuum well points. The wind-powered generation system has been found to provide effective power with a 63.5 cm Hg vacuum being developed in the well point system and a flow of approximately 3.7 ℓ/hr from each well point.

5A11

This work unit, carried out in February 1976, was a small-scale demonstration of hydraulically fracturing in-place dredged material by pressure injection of sand grout. Fracturing at foundation level was expected to produce horizontal fans that would, in effect, produce underdrainage layers in existing masses of fine-grained dredged material. The resulting drainage layers could then be pumped, providing both water table drawdown and vacuum consolidation in the dredged material mass. The field demonstration was a success: 2.5 cu m of grout were placed under a 23- by 15-m area and horizontal grout fans were produced. Pumping tests on the injected sand slurry with 63.5 cm Hg vacuum yielded flows of 11.5 ℓ/hr per well point as compared to 0.6 ℓ/hr from conventional sanded well points installed for comparison. Only 0.5 m excess head of sand grout was needed to fracture the material at the 2.5-m depth. Consequently, only a pipe and funnel were needed to inject the grout rather than the conventional pressure grouting equipment. Additional pumping of the sand-injected layers is currently underway.

5A14

This study is concerned with improving the drying rate of dredged material slurry by periodically mixing the surface crust with the underlying soft dredged material, allowing the crust to reform, then remixing, etc. The process should not be confused with the technique described in Work Unit 5A01. The process is not unlike mechanical stabilization of a highly plastic clay by introduction of coarse-grained cohesionless material. This study has been underway since February 1976. Preliminary data indicate that the periodically mixed area is drying about 15 percent faster than an adjacent control area.

5A15

This work unit is designed to evaluate the effects of conventional hydraulic drainage techniques (i.e., sand layers with collector pipe) for dewatering fine-grained dredged material slurry. Five miniature diked areas are being constructed on the stable dredged sand at the southeast corner of the disposal area. After underdrainage layers are placed, highly plastic organic clay dredged material slurry will be pumped into the test areas and the rate of dewatering and drying will be evaluated. One area will be used as a control section while the other four areas will be used to evaluate the concepts of simple gravity underdrainage, vacuum-assisted underdrainage, seepage consolidation, and seepage consolidation with vacuum assistance.

5A16

This study is the follow-on to the laboratory studies of long-term low voltage gradient electro-osmotic dewatering conducted under 5A04. Although the laboratory studies indicated electro-osmosis was apparently technically and economically feasible, the field demonstration will provide valuable operational data so that the process can be refined under realistic conditions.

5A18

This study is designed to investigate the use of vegetation in providing desired engineering characteristics in disposal areas. Engineering criteria to be satisfied by the vegetation include the ability to dewater the surface of dredged material slurry rapidly and the ability to form a root mat rapidly that is capable of supporting men and equipment.

The use of vegetation for dewatering dredged material is also being evaluated in a field study on Grassy Island in the Detroit River. In this test the common reed *Phragmites communis* was planted prior to placement of the dredged material and allowed to grow. The dredged material was placed so that the plants are almost completely inundated. The plants will send out roots or rhizomes up and down the stalk, dewatering the dredged material throughout the total depth.

2C09

Although this work unit is under Task 2C, the field evaluations of equipment are being conducted at the UPB site. Results of this work unit are directly applicable to work in Task 5A because of the equipment demands for the systems being evaluated. This work unit is concerned with selection and evaluation of both experimental and conventional equipment to perform trenching, earthmoving, and survey and reconnaissance tasks in confined disposal areas. Equipment to be evaluated include low-ground-pressure vehicles, such as the NUC and other military personnel carriers, as well as marsh buggies, backhoes, gradalls, draglines, and dozers, with and without special tracks and operating on and off of matting.

TREATMENT OF CONTAMINATED DREDGED MATERIAL

Background

Investigations of treatment methods for contaminated dredged material are being conducted within the DMRP Task 6B (Treatment of Contaminated Dredged Material). Work within this task includes treatment schemes for dredged material discharged in open-water and confined in diked containment areas. Work units of Task 6B are shown in Table 3.

Preliminary and Laboratory Investigations

6B01, 6B02

Two preliminary studies were conducted to examine the adequacy of certain parameters commonly used to describe the pollution potential of dredged material, to assess conventional treatment methods, and to recommend applicable treatment schemes.

TABLE 3: TREATMENT OF CONTAMINATED DREDGED MATERIAL STUDIES

Work Units	Contractor
6B01 Assessment of Chemical, Physical and Biological Processes for Treatment of Dredged Material	JBF Scientific Corporation, Wilmington, Massachusetts
6B02 Laboratory Treatability studies of Polluted Dredged Material	EEL*
6B05 An Evaluation of Oil and Grease Contamination Associated with Dredged Material	Engineering Science, Incorporated Austin, Texas
6B06 Oxygenation of Dredged Material	JBF Scientific Corporation, Wilmington, Massachusetts
6B07 Flocculation as a Means for Improving Effluent from Confined Disposal Sites	University of Southern California Los Angeles, California
6B08 Development of Design Procedures for Flocculation of Dredged Material	EEL
6B09 Field Evaluation of Vegetation for Removal of Contaminants from Effluent from Confined Disposal Areas	Savannah District (H. L. Windom, Consultant)

* Waterways Experiment Station Environmental Effects Laboratory

Both studies showed that bulk analysis methods of evaluating the characteristics of dredged material do not adequately assess the environmental effects of dredged material disposal. The studies also indicated that many conventional wastewater techniques are frequently inapplicable and/or impractical because the dredged material slurry generally presents a relatively high solids content, a high magnitude and variability of flow, and a complex makeup of physical and chemical properties and organic matter. The studies demonstrated that biological treatment techniques are ineffective, but that chemical coagulation treatment procedure could be employed to reduce suspended solid and the attached contaminants.

Rapid oxygen depletion in the water column is one of the most documented and noticeable effects of open-water disposal of dredged material. During the laboratory studies it was shown that depletion of dissolved oxygen (DO) can be appreciably reduced by aerating the slurry. Both studies recommended field tests to evaluate the feasibility of injecting air or oxygen into the pipeline to reduce the oxygen depletion within the water column. Field tests were conducted and they will be discussed subsequently.

6B05

A study was initiated to investigate problems of oil and grease associated with confined disposal areas. Preliminary results indicate that oil and grease contamination does not appear to be a significant problem. Oil and grease generally are not released from the sediment during the dredging process. The second phase of this study was designed to develop treatment processes for the removal of oil and grease from disposal area effluents. Since the field survey results indicate that oil and grease concentrations in the effluent are so low that treatment will probably not be required, the scope of this phase was changed. Now, bench-scale studies will be conducted to confirm the field findings and to determine factors that may affect oil and grease release.

6B07, 6B08

The use of flocculants to improve the quality of effluent from confined disposal sites is being investigated under these work units. Most contaminants appear to be tied up with suspended solids in the effluent. Consequently, removal of the suspended solids should improve the quality of the effluent. Work being conducted under Work Unit 6B07 is investigating dosages of flocculants needed and the resulting quality of the effluent. Work under Work Unit 6B08 is developing methods for implementing the results of 6B07 under field conditions. Both of these work units are currently active.

Field Tests

Two major field tests have been or will be conducted as part of Task 6B: one to investigate injection of air and oxygen into dredge pipelines to reduce the immediate oxygen demand during open-water disposal of dredged material and the other to evaluate the effectiveness of vegetation in removing contaminants from dredged material in confined disposal areas and thus to improve the effluent water quality.

6B06

Field tests were conducted to evaluate the effectiveness of injecting oxygen and air into the pipeline during open-water disposal operations. Oxygen was injected into the pipeline of a 41-cm dredge while air was injected into the pipeline of a 61-cm dredge. The tests using air were conducted in December of 1975 and the tests with oxygen were just completed.

The oxygenation tests were conducted in Apalachicola Bay, Florida. The water in the area was about 1.8 m deep with a natural dissolved oxygen (DO) value of about 10 mg/ℓ. During disposal operations the DO was depressed to 1 to 2 mg/ℓ near the bottom and 3 to 4 mg/ℓ near the surface and at mid-depth. When oxygen was injected into the system, the DO near the bottom and at mid-depth was increased to about 5 mg/ℓ. No significant change was noted at near surface. Data from the aeration tests conducted in Mobile Bay, Alabama, are currently being analyzed.

6B09

Field tests are currently in progress to evaluate the ability of vegetation to remove contaminants from dredged material and thus improve the water quality of effluents from containment areas. The tests are being conducted at an active disposal site in Savannah, Georgia. The tests are being performed to assess the capability of *Spartina alterniflora* to remove nutrients and heavy metals from the effluents. The site consists of twelve 3-m-wide and 100-m-long runways within the grass. The effectiveness of the grass for removing contaminants will be monitored at various flow rates and distances from the discharge point. These tests will provide general information on the concept. It is realized that more controlled tests will be necessary using various types of vegetation and chemical compositions of effluent to provide a complete design methodology.

CONCLUDING REMARKS

The preceding discussions of research being conducted as part of the Disposal Operations Project have been general with few details given. Since research is in progress, definitive information is not yet available on all aspects of the work. This information is coming in at a high rate and will be analyzed and dissiminated as soon as possible. Summaries of the results of DMRP studies are usually published in the DMRP Information Exchange Bulletin several months before the final report is published. All studies will be completed and final results published in mid-1978.

ECOLOGICAL CONSIDERATIONS IN SITE ASSESSMENT FOR DREDGING AND SPOILING ACTIVITIES

Donald K. Phelps and Allen L. Myers
Environmental Research Laboratory
U.S. Environmental Protection Agency
South Ferry Road
Narragansett, Rhode Island 02882

INTRODUCTION

Dredged materials may contain toxic substances whose effects upon the marine ecosystem are poorly understood. Does ocean disposal of such material have adverse environmental impact? If so, is the degree of impact great enough to justify additional economic burdens that result from:

1) Moratoria on ocean disposal of dredged material resulting from legal actions on the part of environmentalists?
2) Developing alternative methods of disposal that may ameliorate or modify negative effects on the total ecosystem?
3) Development of new or improved technology for the disposing of spoils containing toxicants into ocean systems?

The development of criteria for establishing the acceptability or not for the disposal of dredged materials from marine systems which contain toxicants resulting from a history of man's activities into clean marine areas is extremely important. Given the diversity of coastal marine and estuarine environments, it is unlikely that any single set of dredging and spoiling site criteria can be developed for direct application to all situations. The problems of concern are potential long-term effects on the bottom-dwelling biota of the dredging and dumping of contaminated materials near or in an uncontaminated area. A basic assumption is that healthy benthic communities--whatever their direct economic benefit to humans--are absolutely necessary for the continued function of coastal and estuarine ecosystems.

It is the purpose of this paper to report on a series of studies that have been underway at the Environmental Research Laboratory, Narragansett, Rhode Island, to provide some specific insight into areas of general concern. The primary focus of the studies has been aimed at trying to identify methods that singly or in combination give the most expeditious yet meaningful demonstration of possible negative environmental impacts resulting from the disposal of contaminated sediments into clean marine systems.

The basic structure of this study is comparison. Comparisons have been made between sediments, faunal diversity, and animal tissue residues between highly contaminated and clean benthic systems. The nature of the area of study is such that a series of field stations along a gradient of benthic systems ranging from highly contaminated to a clean system proved helpful.

Animals and sediments have been brought from the field into the laboratory for a series of comparative analyses. The nature of flux from each sediment extreme was measured in a "neutral" water column provided by the seawater system in the laboratory. Responses of the same animal species from each extreme to additional laboratory stress has formed the basis of the development of comparative bioassay as a means for measuring potential impact of contaminants in benthic systems.

AREA OF STUDY

The primary focus of this study is Narragansett Bay, Rhode Island (Figure 1). The Bay has been described 'abnormally stressed' in its upper reaches and as being divisible into a polluted upper Bay, a transitional zone, and a lower Bay having water of high quality. Bottom water salinities range from 28-31 $^{0}/_{00}$ in the upper reaches and from 30-32 $^{0}/_{00}$ in the lower Bay. Temperature escalates from freezing to 22°C throughout the estuary.

The major environmental difference within the system is due to a history of pollution effects in the upper reaches of the Bay. Sources of pollution are domestic waste treatment plants and industrial effluents including metal plating and jewelry manufacturing activities.

Fine sediments in the upper area are anaerobic, characteristically having their redox boundary at the sediment-water interface and having a strong odor of H_2S. In the lower Bay, fine sediments have a well-defined aerobic layer with a redox boundary defined between 50 and 10 cm below the sediment-water interface. Metals in upper Bay sediments include elevated levels of Zn (250 ppm) and Cr (241-363 ppm) compared to lower Bay levels of 12-20 ppm Zn and 57 ppm Cr, respectively. High concentrations of hydrocarbons have been reported in upper Bay sediments compared to levels found in the lower Bay. The upper reaches of the Bay are dredged periodically to maintain shipping channels for the port of Providence.

SUMMARY AND DISCUSSION OF RESULTS

A transect of stations progressing from contaminated to clean benthic systems was studied in Narragansett Bay, Rhode Island (Figure 1). Sediment box cores collected from either end of the transect were brought into a "neutral" water column provided by the flow-through seawater system and held for four months with flow-through rate of 4 liters/min.

Flux of ammonia and metals from the box cores were monitored in the laboratory. Fluxes observed from the contaminated sediments continued to be significantly higher than those observed from uncontaminated sediments (Tables 1-6).

These results demonstrate that a contaminated sediment disposed of in a clean environment will tend to degrade quality of the overlying water column.

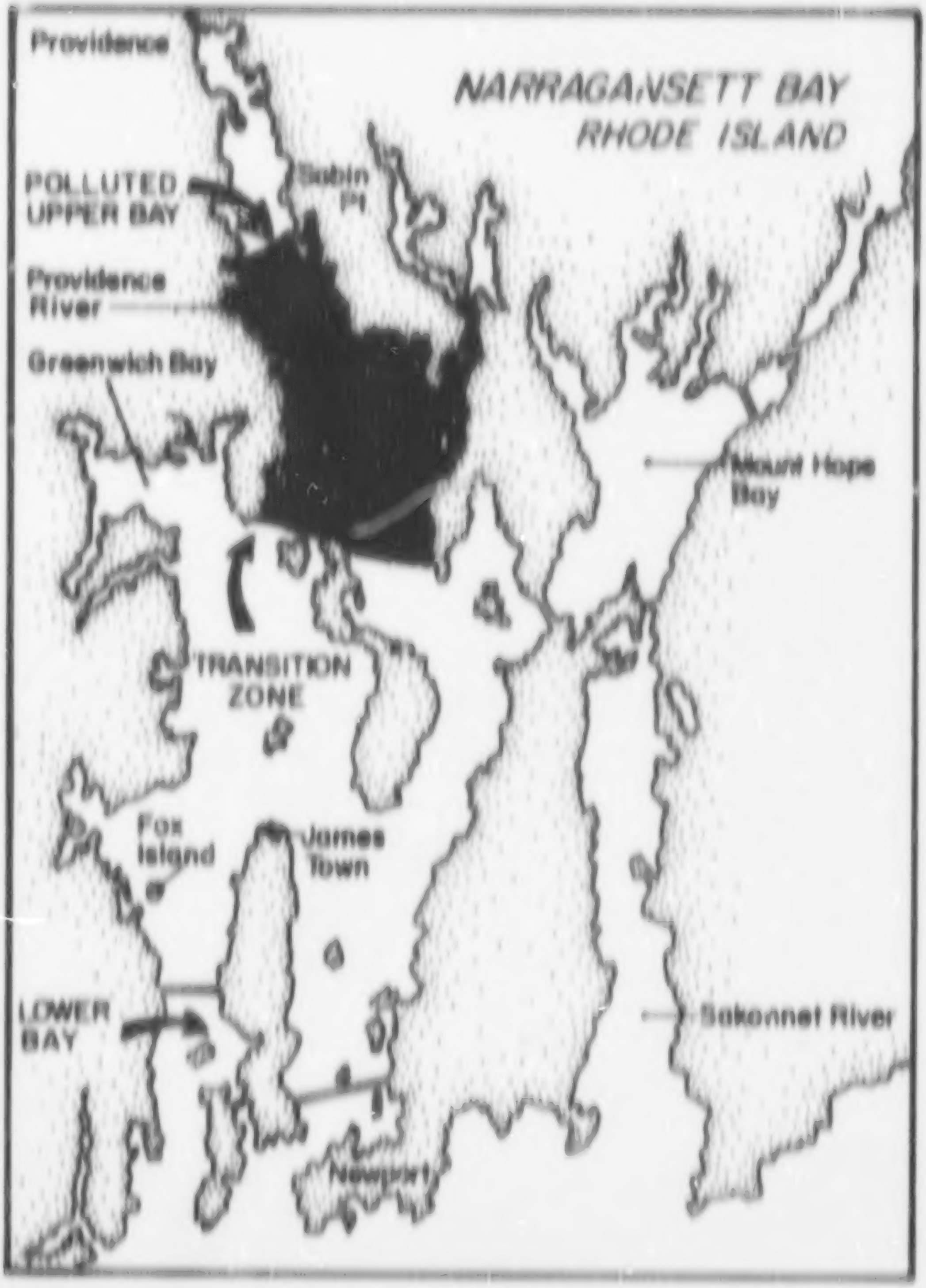

NARRAGANSETT BAY
RHODE ISLAND
Providence
POLLUTED UPPER BAY
Sabin Pt
Providence River
Greenwich Bay
Mount Hope Bay
TRANSITION ZONE
Fox Island
James Town
LOWER BAY
Sakonnet River
Newport

Figure 1. Area of study.

TABLE 1. FLUX RATES OBSERVED FROM LABORATORY-HELD BOX CORES NH_3 IN MICRO MOLES/CM^2/DAY

Dates 1976	Temp. °C	Sabin Point	Jamestown	Ratio Sabin Point: Jamestown
1/7	2.8	0.10	0.01	10
3/31	6.6	0.20	0.09	2.22
4/20	11.9	0.35	0.08	4.38
4/21	21.3*	0.93	0.38	2.45

TABLE 2. FLUX RATES OBSERVED FROM LABORATORY-HELD BOX CORES BOD IN MG/CM^2/DAY

Dates 1976	Temp. °C	Sabin Point	Jamestown	Ratio Sabin Point: Jamestown
3/2	5.4	0.07	0.05	1.20
4/8	8.5	0.073	0.054	1.33
2/9	10.3*	0.142	0.145	.98
4/8	20.2*	0.289	0.174	1.66

TABLE 3. FLUX RATES OBSERVED FROM LABORATORY-HELD BOX CORES MANGANESE IN $\mu G/CM^2$/DAY

Dates 1976	Temp. °C	Sabin Point	Jamestown	Ratio Sabin Point: Jamestown
3/2	5.4	.214	.178	1.20
4/8	8.5	.292	.026	11.64
2/9	10.3*	.378	0.148	2.56
4/8	20.2*	1.542	0.406	3.79

TABLE 4. FLUX RATES OBSERVED FROM LABORATORY-HELD BOX CORES CADMIUM IN μG/CM^2/DAY

Dates 1976	Temp. °C	Sabin Point	Jamestown	Ratio Sabin Point: Jamestown
3/2	5.4	0.031	0.052	0.59
4/8	8.5	0.033	0.024	1.40
2/9	10.3*	0.023	0.036	0.66
4/8	20.2*	0.059	0.016	3.64

TABLE 5. FLUX RATES OBSERVED FROM LABORATORY-HELD BOX CORES ZINC IN μG/CM^2/DAY

Dates 1976	Temp. °C	Sabin Point	Jamestown	Ratio Sabin Point: Jamestown
3/2	5.4	0.591	0.641	0.92
4/8	8.5	0.289	0.300	0.96
2/9	10.3*	0.515	0.098	5.27
4/8	20.2*	0.407	0.240	1.70

TABLE 6. FLUX RATES OBSERVED FROM LABORATORY-HELD BOX CORES COPPER IN μG/CM^2/DAY

Dates 1976	Temp. °C	Sabin Point	Jamestown	Ratio Sabin Point: Jamestown
3/2	5.4	*****	*****	****
4/8	8.5	0.017	0.009	1.95
2/9	10.3*	-0.153	-0.133	-1.16
4/8	20.2*	0.027	0.029	0.95

While dilutional effects may tend to render this degradation insignificant at an undefined distance above the sediment surface, such a flux would exert a significant negative impact at the sediment-water interface.

It may be inferred that the benthic system, once contaminated, provides a major reservoir of contaminants which continue to be exchanged with the overlying water column. A feedback loop of major environmental significance is thereby identified. Regardless of the initial route of transport, whether the media is the water column itself or particulate and/or colloidal scavenging within the water column, the benthic system appears to be the reservoir of greater contaminant concentration. This point is basic in the use of biota as environmental integrators and indicators or predictors. In order to use biological magnification by biota effectively as a *de facto* demonstration of the biological availability of contaminants present in dredge spoils, it must first be established that the sediments are the more significant reservoir and provide a media of biological transport for those contaminants into the overlying water column at a proposed dredge site.

Tissue residue analyses, or total body burden measurements, for some trace metals demonstrate that the levels of metals accumulated by benthic macrofauna and the degree to which the total number of animals within a given population exhibit such accumulation appear to vary as a function of the vertical position in the water column-sediment profile at which the animals feed. Macrobenthic fauna were collected along a transect within the contaminated sedimentary system of upper Narragansett Bay at one point in time.

Fauna were grouped according to the vertical position within the sediment-water profile from which they feed. *Mercenaria mercenaria*, *Mulinia lateralis*, *Ensis directus*, and *Mya arenaria* were included in a group classified as "Water Column Feeders" (WCF) which secure purchase on or in the substrate but feed from the overlying water column. The WCF are all molluscs in this instance.

Two polychaetous annelids, *Nephtys incisa* and *Glycera dibranchiata*, compose the "Subsurface Feeders" (SF). Members of this group actively burrow and feed below the sediment-water interface.

The third and final group is composed of fauna which feed directly from the sediment-water interface or the "Interface Feeders" (IF). The IF's include a polychaete worm, *Pherusa affinis*; two crustacea, *Palaemonetes pugio* (the grass shrimp) and *Rithropanopeus harisii* (the mud crab); as well as two molluscs, *Yoldia limatula* and *Nassarius obsoletus*.

Chromium (Cr), silver (Ag), and zinc (Zn) were measured in total body tissue (molluscs minus their shells) for each animal in each group using neutron activation analysis.

The metal analyses yielded two sets of information (as shown in Figure 2):

1) The percent of total number of animals (from each group) having detectable levels; and
2) a comparison of relative metal accumulation between the groups.

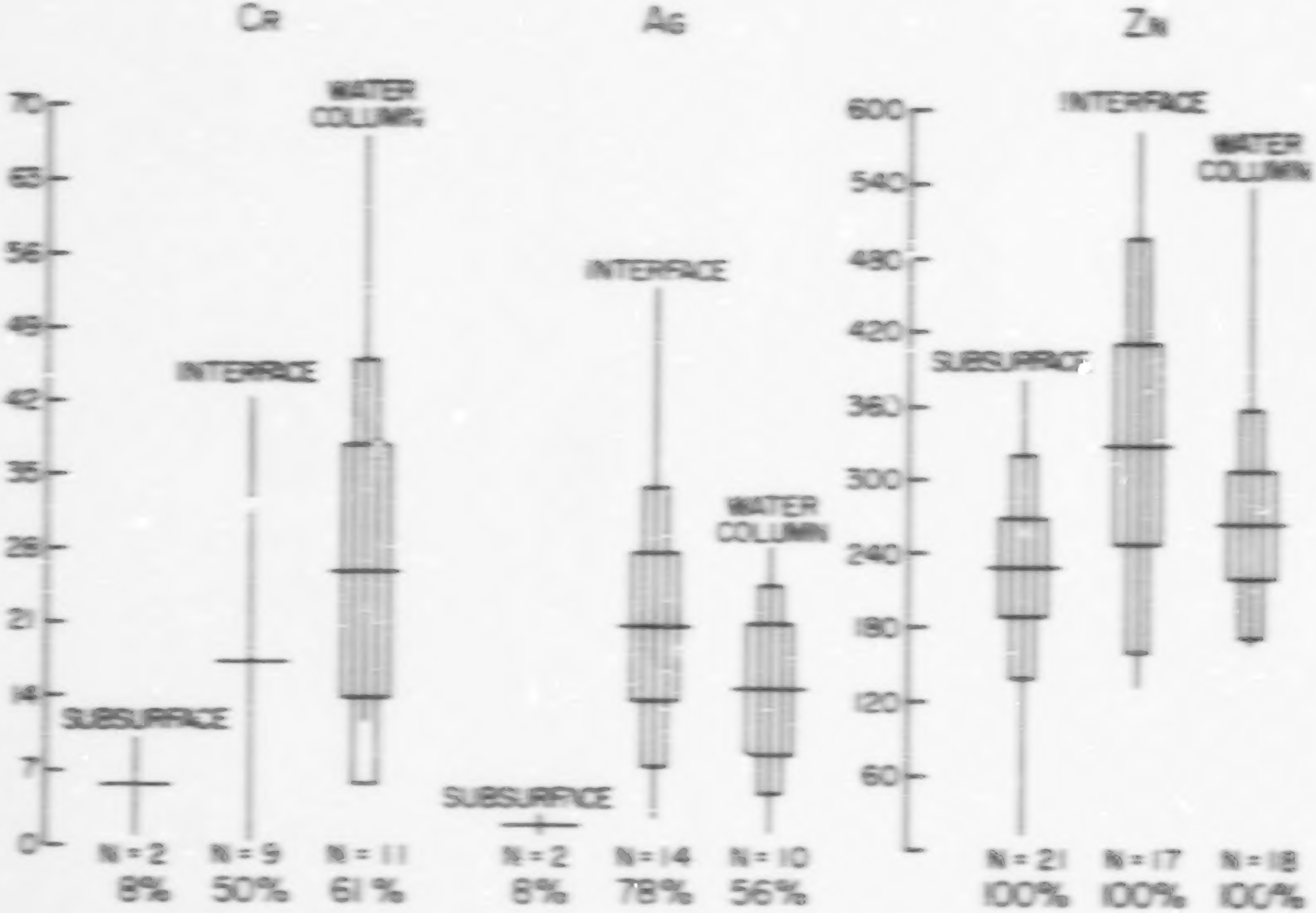


Figure 2. Levels and incidence of occurrence of chromium, silver, and zinc in benthic fauna grouped according to feeding positions.

Of the three animal groups, the SF's had the lowest accumulation of the three metals. Cr and Ag analyses yielded only an 8% incidence of detection in the SF group. This indicates that the group is the least useful in terms of biological integrator or monitoring of contaminated benthic systems.

The WCF's showed the highest level of incidence within the population monitored (61%) and the highest levels of accumulation for Cr. This group showed lower levels of incidence in the case of silver (56%) and accumulation for both silver and Zn than did the IF's.

The IF's showed a 70% level of incidence for Ag and the highest level of accumulation for both Ag and Zn.

All three groups showed a 100% level of incidence for Zn.

These data indicate that the IF's are the most useful group in terms of biological integration and hence monitoring for those contaminants in dredge spoils which are biologically available. This fact takes on greater significance in view of the results from laboratory box core studies described above. Measurable degradation of the water column at the sediment-water interface resulting from the flux of NH_3 and metals through that interface was demonstrated.

WCR's are the better apparent choice for Cr. These results also indicate that they would be useful as integrator-monitors for Ag and Zn as well, but less effective than IF's.

Field observations indicate that, in the contaminated benthic systems of Narragansett Bay, dominance of feeding types shifts in numbers and biomass to the WCF's at the expense of the IF's and SF's which dominate the clean benthic systems of lower Narragansett Bay. This observation suggests that while the IF's may be the best monitor, the group may also be quite sensitive to effects of materials being released from contaminated sediments.

The reduction in numbers of IF's and SF's has a marked effect on the physical system. These animals, through their active burrowing and/or feeding activity, mix the sediments mechanically. Such mechanical mixing contributes to aerating surface sediments. As the animals and their mechanical mixing activity decrease, the redox potential discontinuity migrates closer to and eventually meets the sediment water interface.

The Rhoads Interface Camera, which takes a picture of a sediment profile including the sediment-water interface, is an excellent piece of remote sensing gear. From these pictures, the depth of the RPD (redox potential discontinuity), sediment penetrability which reflects animal activity such as reworking, animal tubes, and burrows can be measured and read. Applied to the transect, the camera allowed us to demonstrate that the RPD approaches the sediment surface as one moves up the bay; further, that the sediment penetrability increases in the same direction. We are currently evaluating other kinds of information which can be inferred from the interface photographs.

A gross measure of animal diversity, expressed simply as the total number of species collected per unit measure of surface area, primarily reflects the reduction in numbers of IF's and SF's referred to above. A transect including six stations ranged from clean areas of Narragansett Bay (stations 1, 2, 3 and 4) into areas having sediments of higher levels of contamination (stations 5 and 6) (Figure 1). Stations 1 and 6 are the stations from which the "clean" and "contaminated" box cores, used in the laboratory flux experiments, were respectively collected.

Total numbers of species are listed for each season (Table 7). Stations 5 and 6, within the Providence River, have much lower numbers of species than stations 4, 3, 2 and 1, which are located outside it. Moreover, though the two end stations, 1 and 6, have only four species in common in the fall, they have 11 species in common the following spring due to an influx of juveniles into both stations. At station 1, some of these juveniles may be killed by trophic group amensalism during which deposit feeders push out the filter feeders. At station 6, observations suggest that most species are killed off by late summer conditions. The fact is established that, for larvae at the settling stage, both stations offer "acceptable" sediment. This material represents only a preliminary report on the data collected.

TABLE 7. NUMBERS OF SPECIES IN QUANTITATIVE INFAUNAL SAMPLES AT EACH OF 6 STATIONS FROM THE PROVIDENCE RIVER (6) TO MID-NARRAGANSETT BAY (1), BY SEASON

	Station					
Season	6	5	4	3	2	1
July 1975	5	10	19	18	19	22
January 1976	12			18		24
June 1976	20	18	29	25	28	30

An initial study of tissue residues on total body burden measurements for cobalt, iron, and zinc was made on *Mercenaria mercenaria* distributed throughout Narragansett Bay. Two statistical populations of this water-column-feeding mollusc were identified on the basis of those levels of metals contained in their tissues (Figure 3). One population, which has significantly higher levels of some metals, occurs from the upper portion of Narragansett Bay and into the Providence River (Figure 1). The statistical population found in the clean lower Bay (Figure 1) is characterized by lower metal levels. Those *M. mercenaria* having higher metal levels are also characterized by the presence of dark pigment which infuses the tissues, as well as significantly higher levels of yellow-staining amoebocytes and development of black concretions in the kidneys as evidenced by histopathological examination. More intensive collections of *M. mercenaria* were made at stations representing areas typical for the occurrence of each statistically identified population. Cabin Point and Fox Island were the stations where collections were made (Figure 1). The metals analyzed were expanded to include chromium (Cr), cadmium (Cd), silver (Ag), copper (Cu), lead (Pb), and nickel (Ni), as well

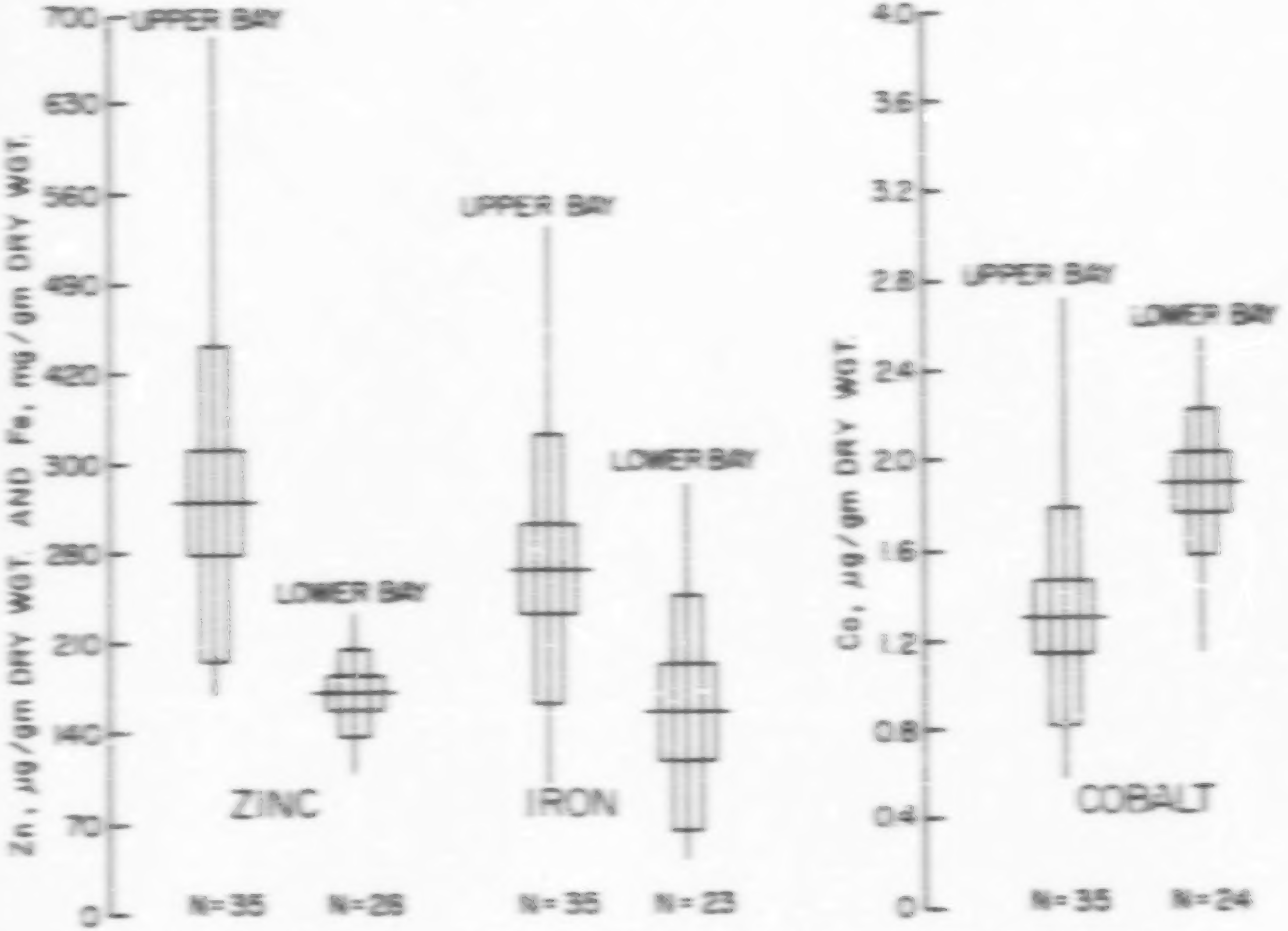


Figure 1. Levels of zinc, iron, and cobalt in grouped samples of *Mercenaria mercenaria* from Narragansett Bay, Rhode Island.

as including zinc (Zn) and cobalt (Co) which were measured in the initial study. Results (Figure 4) indicate significantly elevated levels of Cr, Zn, Cd, Cu, Pb, and Ni in *M. mercenaria* collected from contaminated sediments, as compared to those collected from clean sediments. No significantly elevated differences were demonstrated between the two *M. mercenaria* populations in the case of Ag and Co.

Two basic questions were generated from these results:

1) Would those *Mercenaria* having higher metal levels depurate those metals if held in clean water for a reasonable time period?
2) If higher levels of metals were retained, do those *M. mercenaria* having higher levels exhibit differences in physiological responses?

Samples of *Mercenaria* having elevated levels of metals and the group having apparently lower were collected from Sabin Point and Fox Island, respectively, and brought into our laboratory seawater system. They were held for a period of 30 days in ambient temperatures of 24°C ± 3°C. At the end of that time, soft tissues were removed from the shells and analyzed by atomic absorption analyses for the metals identified above. Ambient temperature levels were optimum for *Mercenaria* pumping and feeding roles.

The results identify three types of conditions in the *Mercenaria* metal relationship:

1) That condition in which metals are retained in *Mercenaria* collected from contaminated ("dirty") sediments at statistically higher levels than in those *Mercenaria* collected from uncontaminated ("clean") sediments after 30 days depuration in a neutral system (Group I, Figure 5). This group includes the metals Cd, Cu, Ni, Pb, and Ti.
2) The second situation is that in which there was no measurable difference in metal levels between the "clean" and "dirty" *Mercenaria* before or after depuration. Metals falling into this group (Group II, Figure 6) include Mn, Zn, V, and Co.
3) The third and final situation is demonstrated by Group III (Figure 7). In this instance metals which were significantly higher in "dirty" *Mercenaria* were in fact depurated to a level comparable to "clean" *Mercenaria* after 30 days. Two metals, Ag and Al, compose this group.

In the case of the Group I metals, the data indicate a strong probability that bioconcentration is taking place. *Mercenaria* are concentrating some potentially toxic metals (Cd and Pb especially) from contaminated sediments. They are making those materials available for biomagnification throughout the food chain. *Mercenaria* are consumed directly by man. Dredging of contaminated sediments and disposing of them in an area where commercial harvesting of *Mercenaria* is taking place could pose a reasonably serious threat to man as a consumer. Copper reached levels in "dirty" *Mercenaria* five times greater than in "clean." Cadmium, Pb, Ni, and Ti levels were three times greater in the "dirty" *Mercenaria*. Group I metals should be given high priority for purposes of monitoring.

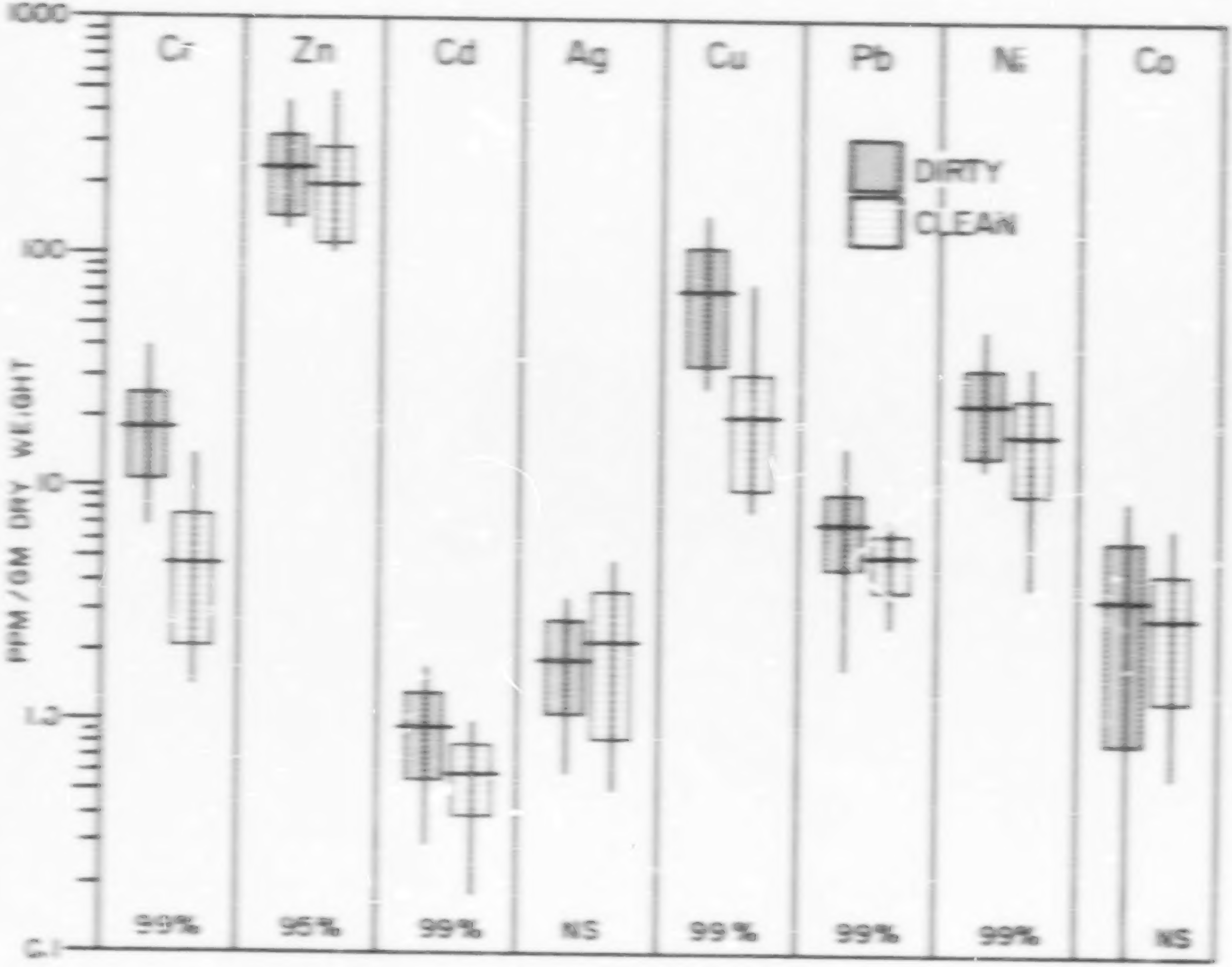


Figure 4. Comparisons of metal levels between *Mercenaria mercenaria* from contaminated ("dirty") and uncontaminated ("clean") sediments from Narragansett Bay, Rhode Island.

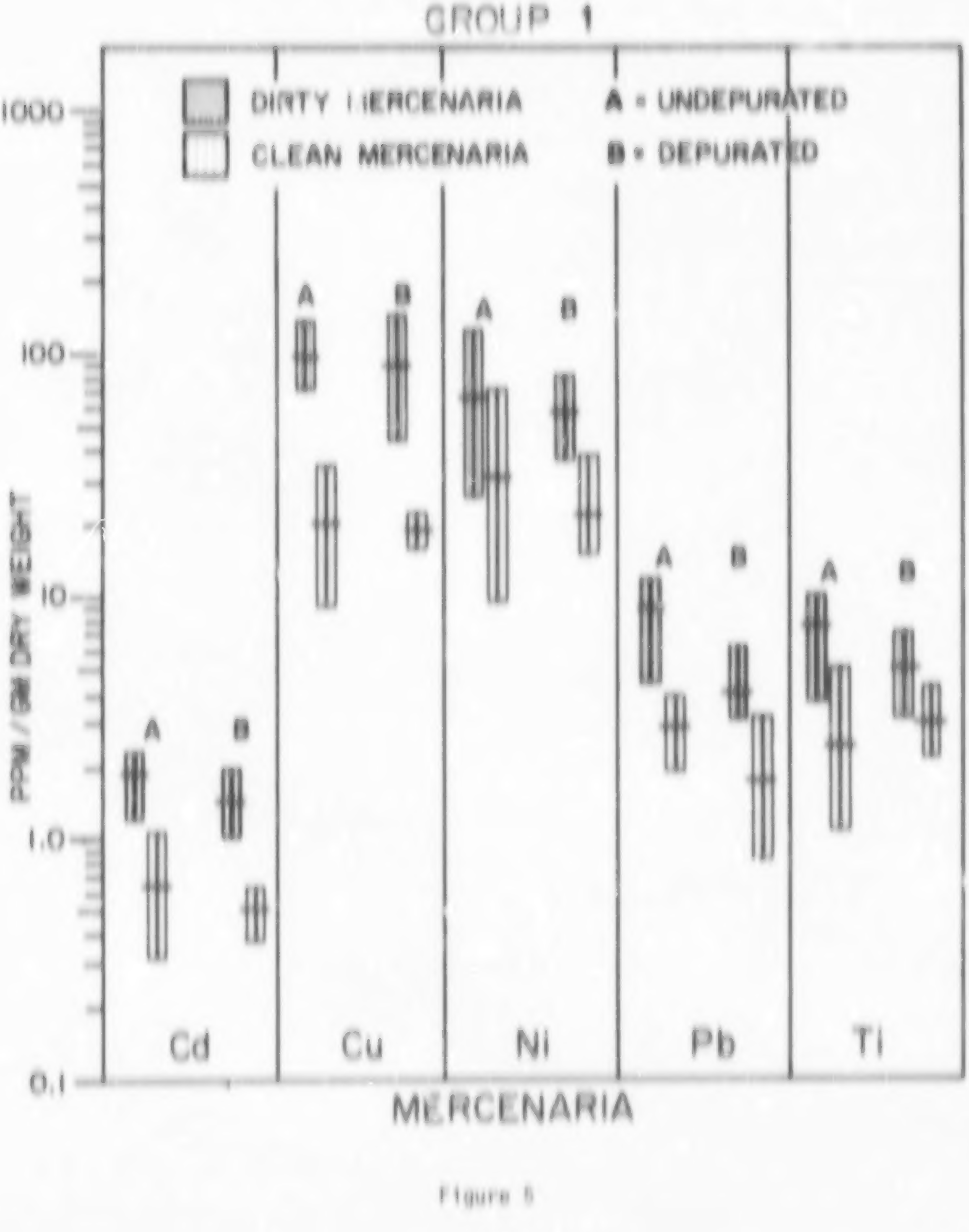

GROUP 1
DIRTY MERCENARIA
CLEAN MERCENARIA
A = UNDEPURATED
B = DEPURATED
PPM / GM DRY WEIGHT
1000
100
10
1.0
0.1
A
B
Cd
Cu
Ni
Pb
Ti
MERCENARIA

Figure 5

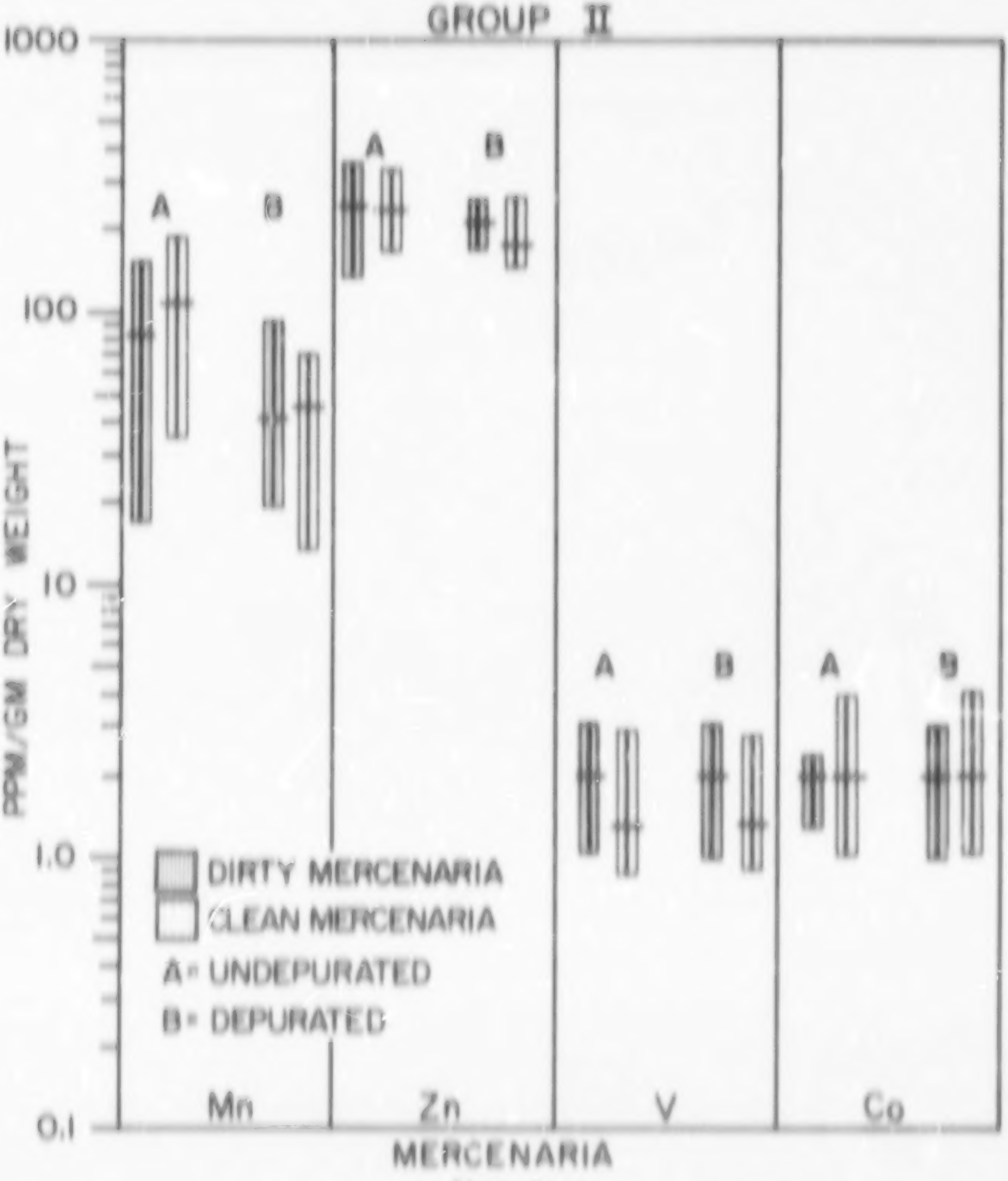

GROUP II
1000
100
10
1.0
0.1
PPM/GM DRY WEIGHT
A
B
Mn
Zn
V
Co
DIRTY MERCENARIA
CLEAN MERCENARIA
A= UNDEPURATED
B= DEPURATED
MERCENARIA

Figure 6

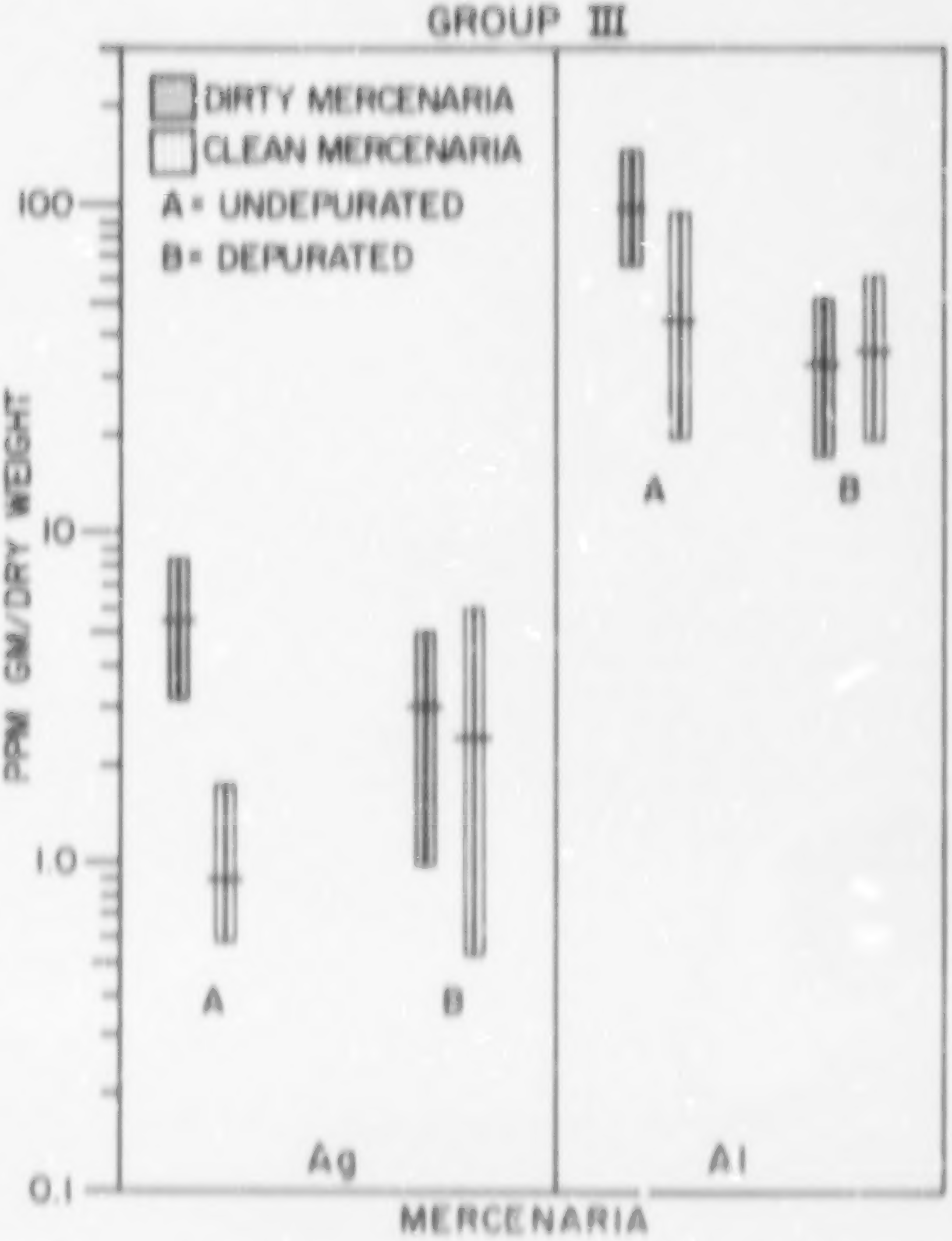

GROUP III
DIRTY MERCENARIA
CLEAN MERCENARIA
A = UNDEPURATED
B = DEPURATED
PPM GM/DRY WEIGHT
100
10
1.0
0.1
A
B
A
B
Ag
Al
MERCENARIA

Figure 7

Of the Group II metals, Mn, Zn, and Co have been detected in significantly higher levels in contaminated upper Bay sediments. The fact that *Mercenaria* do not reflect such elevated levels in their tissues may be due to the fact that the metals are fluxed from the sediments (as demonstrated in the case of Mn and Zn) in a state that may render them biologically unavailable or that those molluscs regulate those metals biologically. No information is available for V levels in sediments. At any rate, Group II metals are of little use for monitoring purposes in this instance.

Group III metals, Ag and Al, depurate from "dirty" *Mercenaria* to lower Bay levels within the 30 day period. Apparently, those metals, when initially detected, were part of the gut content and were subsequently eliminated. Monitoring for these metals may give insight into movement from disposal sites. While they may not be biologically available, their presence in the *Mercenaria* indicate movement through or from a system of greater concentration to lesser concentration and probably in a particulate state.

Mercenaria were collected from the same upper and lower Bay stations described previously to conduct another experiment designed to see if the high metal and low groups responded differently.

Metal uptake under laboratory conditions was decided upon as a reasonable measure for response. A wide range of reactions are involved in the uptake of metals. Simple processes such as valve opening and closing and through complex biochemical exchange reactions are bracketed in such an uptake study.

Clams from each location were placed in a series of tanks having radioactive ^{65}Zn alone, ^{65}Zn plus Ag, and ^{65}Zn plus Cu. Three clams were taken from each group and ^{65}Zn measured and compared at the end of 2, 4, 8, and 16 days, respectively.

Results are as follows:

The uptake pattern for Zn alone was very different between the two groups. "Dirty" clams had taken up more than twice as much of the ^{65}Zn by the end of the experiment (Figure 8).

Animals exposed to the combination of ^{65}Zn and silver demonstrated a basic similarity in the uptake pattern; but "dirty" clams had a higher uptake of ^{65}Zn at the end of the experiment also (Figure 9).

The most dramatic difference was in the exposure of ^{65}Zn in combination with Cu. While uptake patterns were somewhat similar, the highest ^{65}Zn uptake level of all occurred in the "dirty" clams of this experiment in the presence of Cu (Figure 10).

The previous depuration experiment demonstrated a real bioconcentration of five times the Cu level in "dirty" over "clean" clams. The next question is--is the different uptake level of ^{65}Zn in the presence of Cu demonstrated in this experiment due to the existing body burden of Cu in the "dirty" experimental animals? This is a question to be answered by further research efforts.

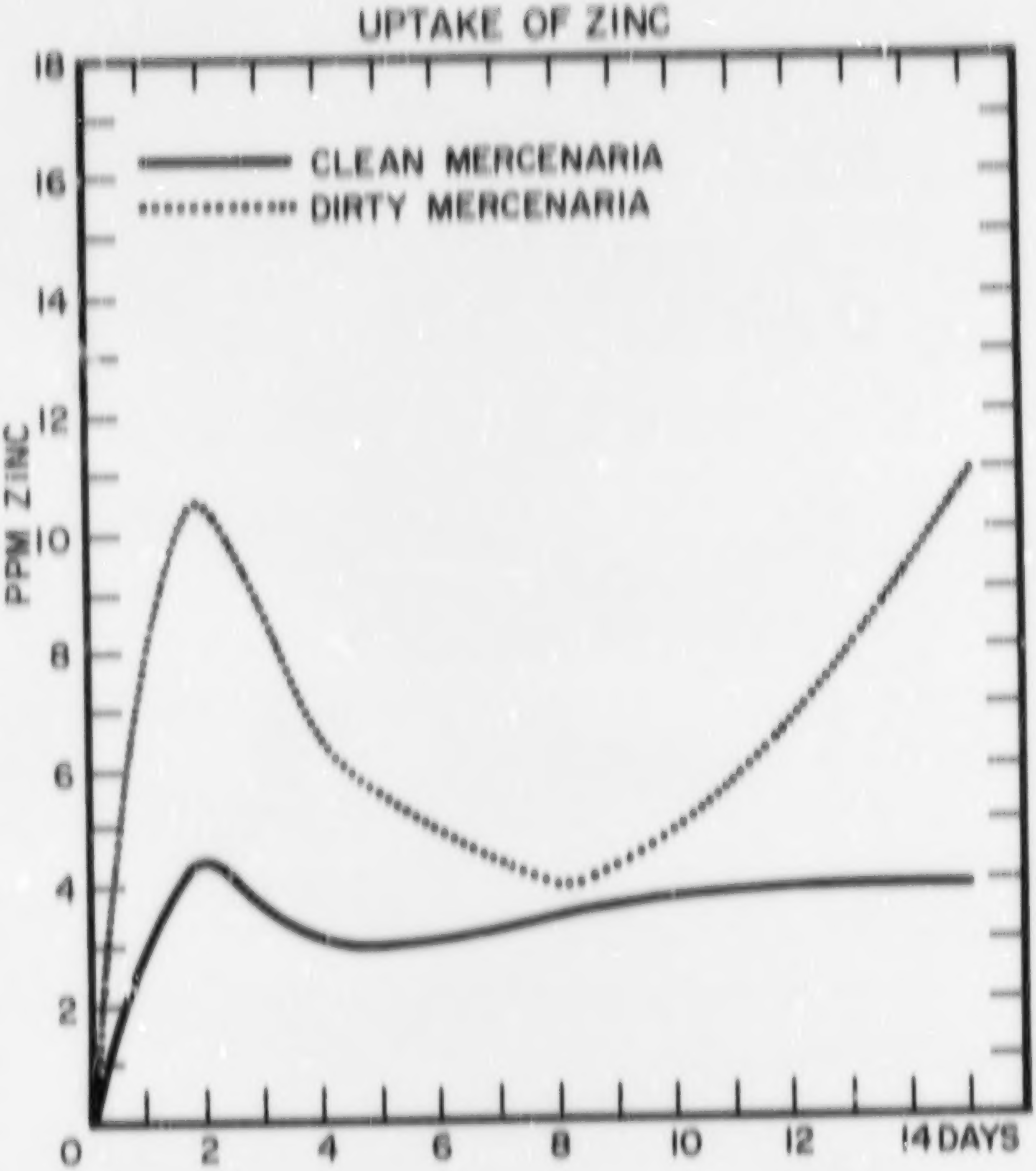
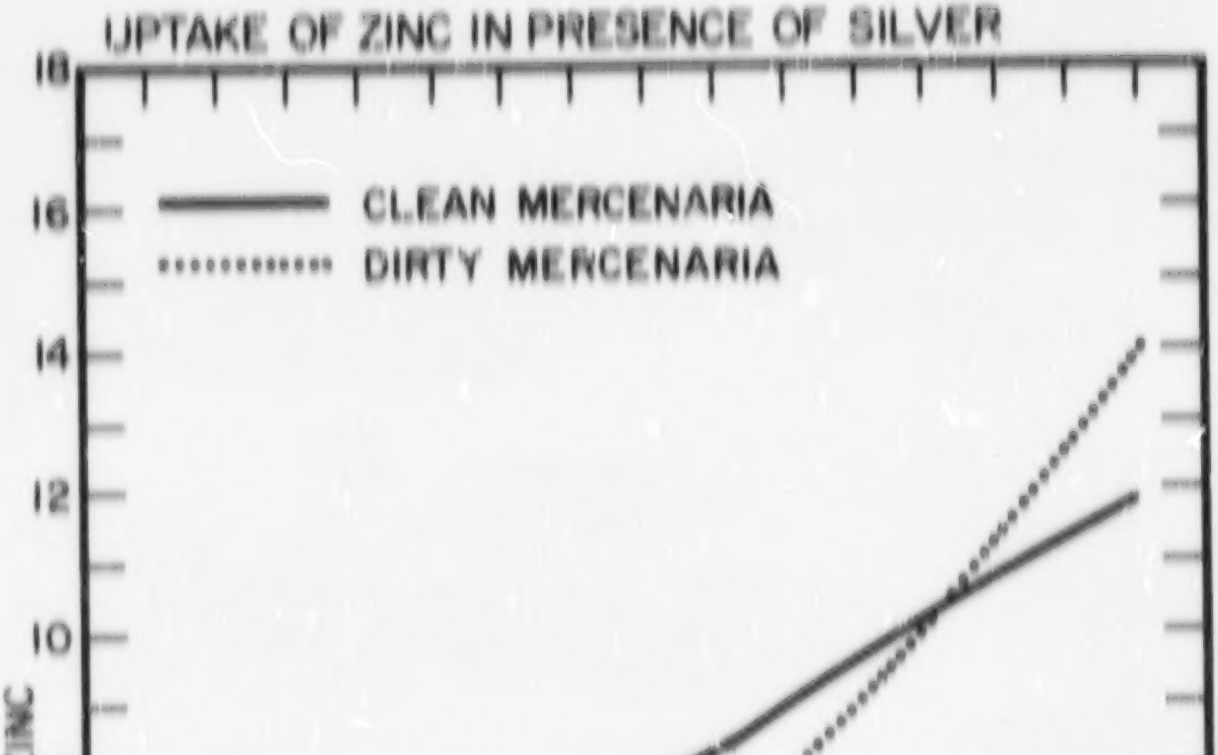

UPTAKE OF ZINC
CLEAN MERCENARIA
DIRTY MERCENARIA
PPM ZINC
18
16
14
12
10
8
6
4
2
0
2
4
6
8
10
12
14 DAYS

Figure 8



UPTAKE OF ZINC IN PRESENCE OF SILVER

Figure 9

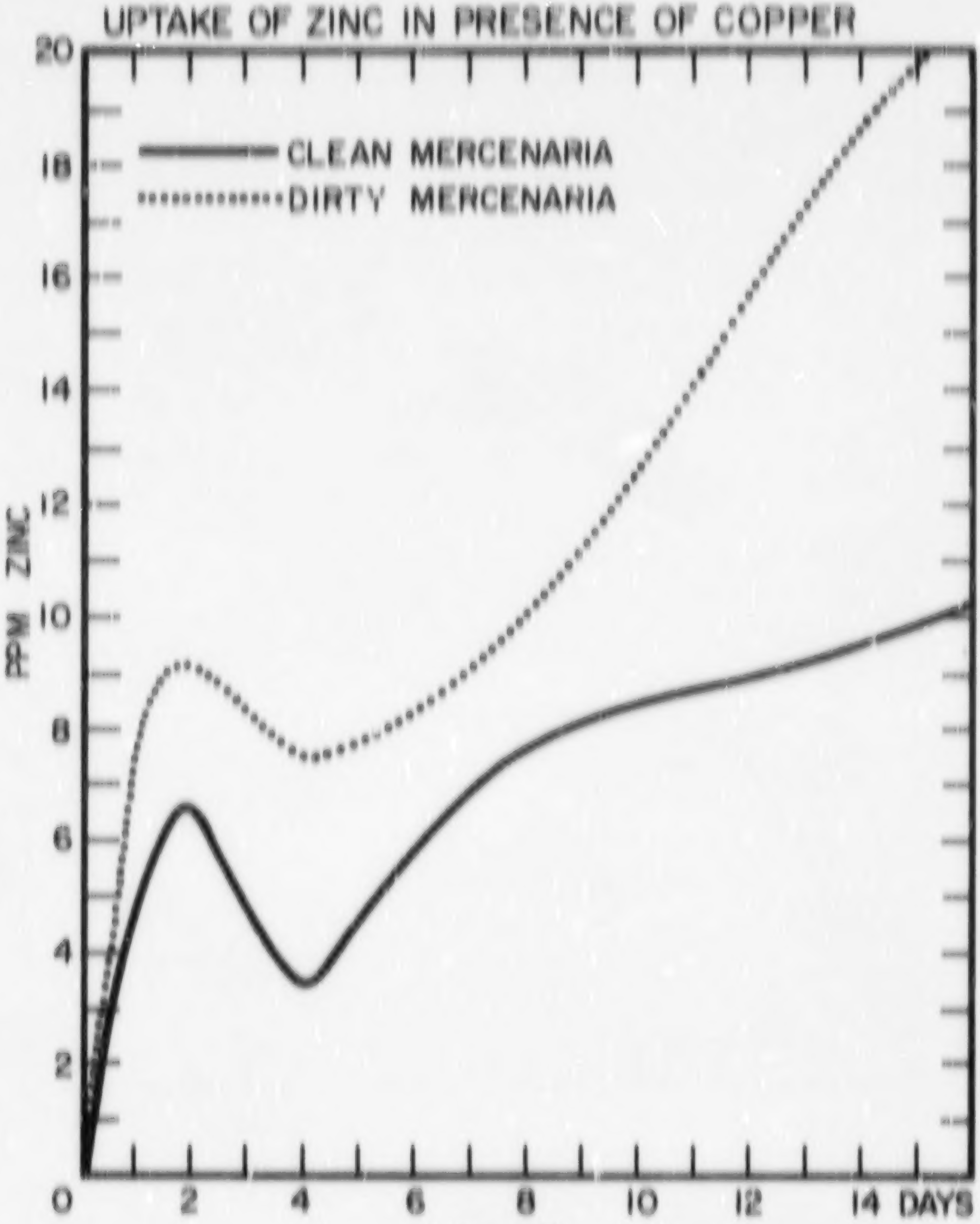

UPTAKE OF ZINC IN PRESENCE OF COPPER
CLEAN MERCENARIA
DIRTY MERCENARIA
PPM ZINC
20
18
16
14
12
10
8
6
4
2
0
2
4
6
8
10
12
14
DAYS

Figure 10

The upper Bay population behaved differently. It may be deduced that the different behavior results from a combination of impacts stemming from man's pollutional contributions to the upper Bay systems.

While the "dirty" clams have higher metal levels, they also have higher hydrocarbon levels. They have demonstrated differences in response and have distinctly different histophathological histories as well. They even look different. Obviously, specific cause and effect mechanisms are well buried in this multi-stressed population.

While *Mercenaria* are WCF's and studies described above indicate that IF's stould be the biological monitors of choice, only *Mercenaria* are ubiquitously distributed throughout the Bay. This fact added to the fact that *Mercenaria* live for periods exceeding two years made them the biological monitors of choice in the preceeding two experiments.

RECOMMENDATIONS

Data presented in this paper demonstrate that metals and other materials (such as NH_3) continue to be fluxed from contaminated sediments upon relocation or redeposition into a "clean" discharge site. Fluxed materials are taken up in various levels and combinations by a variety of macrobenthic fauna. The position at which fauna feed and ventilate (in a vertical profile through the sediment-water interface) is related to the kinds and amounts of materials which are accumualted by them.

Mercenaria having significantly higher levels of certain metals in their tissues demonstrate different uptake patterns and levels when exposed to additional amounts of those same metals in the laboratory under a parallel bioassay run simultaneously with *Mercenaria* having significantly lower amounts of metals. This fact provides an initial indication that response mechanisms differ between the same species (*Mercenaria* in this case) living in contaminated and uncontaminated benthic systems.

It is recognized that while *Mercenaria* having higher tissue levels of metals respond differently from the same species having lower tissue residues, no cause and effect relationship has been established.

It is therefore recommended that:

1. Heavy metal tissue elevations resulting from exposure to contaminated sediments be examined for use as a tag for a variety of possible stressors coming from contaminated sediments. For example, it is known that *Mercenaria* having higher tissue residues of metals have a variety of other problems that are associated with contaminated sediments. These include elevated levels of total petrochemical hydrocarbons (reported in the literature), histopathological, parasitological, and biochemical abnormalities. However, for purposes of monitoring, good metal analyses are more reliably carried out at much lower cost in currency and expertise than are analyses for petrochemicals and other organics of interest or than are sophisticated histopathological, parasitological, or biochemical analyses. Another profitable avenue for research may be the examin-

ation for possible direct relationships between metal levels and the presence and possible quantity of other contaminants in the tissue of fauna living in contaminated sediments.

2. Parallel bioassays should be run between fauna collected from sediments that are of questionable and known quality before those sediments be disposed of in clean systems. The parallel bioassay can be used as a direct demonstration of possible negative environmental impact on biota from contaminated sediments. Other types of responses than the metal uptake approach, used in this study, should be examined. For example, we are currently looking for differences in feeding rates and carbon fixation between *Mercenaria* from contaminated and uncontaminated sediments. We intend to examine different responses that additional stress may elicit from these parallel populations. We believe that fauna living in contaminated sediments provide valuable insight into the effects that those sediments will have on similar fauna living in a "clean" site that may be proposed for dredge disposal. A direct demonstration of possible negative environmental impact due to disposal of contaminated sediments may be outlined by a series of judicious comparisons made on the basis of biological responses and tissue residues.

APPENDIX A

Notes on the units of measure and methodology used by Japanese authors.

(1) Taisuke Sameshima--	Concentrations of toxic substances in sediment reported in the original manuscript as ppm were converted to mg/kg (dry weight).
(2) Tamonri Ohtsuka--	Concentrations of toxic substances in sea water reported in the original manuscript as ppm were converted to mg/l and the concentrations of toxicants in sediment originally reported as ppm were converted to mg/kg (dry weight).
(3) Akio Murakami--	The unit "SS" refers to suspended solids or suspended matter (See Appendix B).
(4) Motoo Fujiki et al.--	It may be assumed that concentrations of toxicants in the muscle tissue of fish and in the bail (prawns) were reported on a net weight basis unless specified otherwise. The "SS" refers to suspended solids (See Appendix B).
(5) Hiroaki Egawa and Shizuko Tajima--	All mercury concentrations in the paper are reported as μ/liter.
(6) Ken Murakami and Kazuo Takeishi--	Units of measurement are those used in the original text except for the "Tentative Standards" units which were reported as ppm and now converted to mg/kg for uniformity.
(7) Tetsuo Toshida and Yoshikazu Ikegaki--	"SS" concentrations reported as ppm (See Appendix B).
(8) Shingo Fujino--	Units of measurement are as reported in the original text except ppm has been changed to mg/kg when concentrations of toxicants in spoils are referred to.
(9) Tatsuro Okumura--	Units of measure are as they were in the original text.
(10) Eisuke Satoh--	"SS" concentrations reported as ppm (See Appendix B).

APPENDIX B

Turbidity and suspended solids go hand in hand and may at times be used somewhat interchangably. The two parameters are, however, quite different and should not be confused. The following defines the two terms and describes how they are measured and reported in this publication according to the Japanese Industrial Standard.

TURBIDITY

Turbidity--	the cloudy condition of water due to the suspension of silt, finely-divided organic matter or other pollutants. In Japan the standard unit of measurement is ppm. One ppm turbidity is defined as the degree of cloudiness present in water containing 1 mg of refined Kaolin in 1 liter of water.

GENERAL METHOD

There are two methods of measuring turbidity. Both measurements are made by comparing the test water with a standard turbidity solution.

Standard Turbidity Solution

Place 1 gram Kaolin in a 1 liter flask, add 10 ml formalin and dilute to 1 liter with water. This is the primary standard solution. Secondary standard turbidity solutions are made by diluting the primary standard, e.g. 1 ml of the primary standard made up to 10 mls would provide a standard solution in which 1 ml of solution would contain 0.1 mg of refined Kaolin.

Measurement

Place 100 ml of test water in a test tube. Place 2, 4, 6, 8, and 10 ml of the secondary standard solution in the same type of test tubes and dilute them to 100 ml with water. Visually compare the turbidity of the test water to the five different standard solutions by looking downward through the length of the test tube. If the turbidity of the test water is the same as one of the tubes which contains A ml of standard turbidity solution, "Turbidity" is calculated as follows:

$$T = A \times \frac{1000}{I} \times 0.1$$

Where T = "Turbidity"
I = Quantity of test water (mls)

note: If the turbidity of the test water exceeds 10 ppm, dilute the test water to less than 10 ppm and use the same method.

PHOTOELECTRIC PHOTOMETER METHOD

To Measure Transmitted Light

This is the method to measure light which passes through the test water at a wavelength of 660 mμ.

To Measure Scattered Light

This is the method to measure light scattered by small particles in the test water.

Both of these methods require a calibration curve to be prepared with standard turbidity solutions in order to determine the actual "turbidity."

SUSPENDED MATTER (Suspended Solids, "SS")

Suspended Matter-- the substance which can be separated by filtration or by means of the centrifugal separator. It shall be determined by any of the following methods. When the test water is difficult to filter, apply the centrifugal separator method, and when the test water contains extraordinarily large quantities of suspended matter, apply the Buchner funnel method. Test water shall be taken from the waste water passing through a 2 mm mesh sieve. The lowest detection level is 5 mg.

FILTRATION THROUGH FILTER PAPER

Sintered Glass Filter Method

1) Apparatus

Sintered Glass Filter--A crucible type glass filter 1G2 or a Buchner funnel type sintered glass filter 3G2.

2) Operation

Prepare two sintered glass filters of the same type and approximately same weight. Lay six sheets of filter paper on each one and pour water over them several times so they adhere together. Then transfer the filters to a circulating air oven and dry them for two hours at 105-110°C. Allow filters to cool in a desiccator, and weigh to the constant weight (when a chemical balance is used, the lighter filter may be used as a supplementary weight). Then pour a suitable amount of the test water into the heavier filter[1], filter by suction, and wash down the substances adhering to the wall of the filter with the filtrate several times. Next, pour the filtrate into the lighter filter several times and filter by suction. Dry the two filters in the air oven for two hours at 105-110°C, and allow to cool in a desiccator. Weigh each filter (when a chemical balance is used, the lighter filter may be used as a supplemental weight), obtain the difference in weight before and after filtration, and calculate the quantity of the suspended matter in ppm according to the following formula:

$$S = (a-b) \times \frac{1000}{V}$$

Where S = Suspended matter (ppm)
a = Difference in weight between before and after the filtration of the test water (mg)
b = Difference in weight between before and after filtration of the filtrate (mg) (when a chemical balance is used, b = 0)
V = Amount of test water (ml)

Remarks 1. When determining the ignition loss of volatile surpended matter, the test shall be carried out in accordance with GFP method specified in Remarks 3, or after washing out the suspended matter together with filter paper into a crucible or an evaporating dish as much as possible, then dry and ignite in the muffle furnace.

Remarks 2. When soluble evaporated residue is less than 5000 ppm, correction, due to the difference in weight of the filtrate before and after filtration, may be omitted. However, when chemical balancc is used, lighter filter shall be used as supplemental weight, so the filtration of the filtrate shall be carried out at the samn time.

Even when the direct reading balance is used, the weight varies with the hygroscopic property of the substances contained in the test water and other conditions, so it is desirable to have the correction performed by obtaining the blank test value of the filter through which the filtrate is passed. When the test water contains a lot of fats and oils, grease, wax, etc., a part of these substances shall be determined as the suspended matter.

---

[1] The water sample must be sufficient to produce a suspended solids weight in excess of 5 mg (the lower detection limit). Ordinarily, 200 ml of test water would be enough. However, for test water which is rather difficult to filter, add 10 ml of test water just before the test water is through filtering. Add the test water when filtration has nearly stopped and use the total amount of added test water as the volume for testing.

When a determination of suspended matter exclusive of oils and fats is required, pour 10 ml of n-hexane over the dried and weighed filter several times to wash out the fats and oils. Then dry the filter and weigh again.

Remarks 3. Glass Fiber Paper Method (GFP Method)
Place on a Nutsche or suitable supporting plate a GFP (Whatman GF/B or similar) filter of known weight, which has been dried at 105 to 110°C for 2 hours after washing. Pour a proper amount of test water into it so the weight of the suspended matter after drying and filtration will exceed 5 mg. Put a portion of the filtrate back in the original test water container. Then wash down the suspended matter adhering to the walls of the container and vacuum filter on GFP again. Repeat this operation several times and remove as much water as possible. Next, detach GFP from the filter and transfer it to a watch glass, etc. Then proceed as described in the discussion on operation of the Sintered Glass Filter Method and obtain ppm of the suspended matter.

After determining the suspended matter, determine the ignition residue in the suspended matter, if necessary. Do this according to the operation described in the section on Sintered Glass Filter Methud.

Buchner Funnel Method

This method is applicable to samples containing a large quantity of suspended matter such as sludge.

1) Apparatus

Perforated Plate Made of stainless steel (SUS 27 or 28). Its size is about 0.5 mm in thickness, about 50 mm or 90 mm in diameter and it can be put in a Buffner funnel to be used. It is shaped like a watch glass with a slightly-bent edge. Small mesh about 0.5 mm in diameter is bored at suitable intervals over the flat surface.

Rubber Packing Rubber packing is a rubber ring about 2 to 3 mm in thickness, about 10 mm to 90 mm in diameter and about 10 mm in width. It can be put in a Buchner funnel and used for filtration by means of suction, putting the perforated plate on it.

Buchner Funnel About 50 mm or about 90 mm.

2) Testing Operation Prepare two perforated plates. Put rubber packing in Buchner funnel and plate.

Place the perforated plate on it. Then lay the filter paper on (Grade 6), pour water through the filter paper several times and apply vacuum. Remove the filter paper with each perforated plate after drying it at 105 to 110°C for 2 to 3 hours. Allow to cool in a desiccator and weigh to the constant weight (when chemical balance is used, lighter perforated plate shall be used as a supplemental weight). Next put the heavier perforated plate and with the filter paper in the funnel and filter 200 to 400 ml of the test water by means of suction. Then pour the filtrate into the lighter plate with filter paper several times and continue the suction. When filtration is completed take out the perforated plate with filter paper on it and dry at 105 to 110°C for 2 to 3 hours. Allow to cool in a desiccator and weigh (when chemical balance is used, the lighter plate shall be used as a supplemental weight).

Obtain the difference in weight before and after this operation, and calculate ppm of the suspended substances contained in the water by the following formula:

$$S = (a - b) \times \frac{1000}{V}$$

Where S = Suspended substances (ppm)
a = Difference in weight before and after filtration of the test water (mg).
b = Difference in the weight before and after filtration of the filtrate (mg). (When chemical balance is used b = 0)
V = Test water (ml).

Remarks: Same as remarks 2, 3.

FILTRATION THROUGH ASBESTOS LAYER

1) Apparatus Gooch crucible. 25 to 35 ml.

2) Reagents Suspension of asbestos. Add water to 15 g of asbestos for Gooch crucible and after removing fine portion by decantation several times, add water to make up to a liter.

3) Operation Prepare two Gooch crucibles (same shape and approximately same weight). After drying, pour about 20 ml of suspended asbestos solution well-stirred to obtain a layer of asbestos about 3 mm thick (about 0.3 g)[2] and gently apply suction. Place crucibles into the air oven. After drying for two hours at 105 to 110°C, allow to cool in a desiccator and measure the weight of each crucible to the constant weight (when chemical balance is used, lighter crucible shall be used as a supplemental weight). Attach the heavier crucible to the suction

---

[2] When half the amount of asbestos solution is poured, place the perforated plate and pour the other half of the solution.

bottle and pour a proper amount (so as to make the suspended matter after drying over 5 mg) of test water into the crucible and gently filter by means of suction. At this time, repeat the filtration of the initial portion of the filtrate.

Next pour a small amount of filtrate through the lighter crucible several times during suction, when the filtration is concluded, place it in the air oven and dry for two hours at 105 to 110°C, and allow to cool in a desiccator. Weigh the crucible, obtain the difference in weight, and calculate ppm of the suspended matter by the following formula:

$$S = (a-b) \times \frac{1000}{V}$$

Where S = Suspended matter (mg)
a = Difference in weight before and after filtration of the test water (mg)
b = Difference in weight before and after filtration of the filtrate (mg). (When chemical balance is used, b = 0).
V = Amount of test water (ml)

Remark: Sampling method of the test water shall be performed in accordance with Footnote 1 specified in glass filter method. When the soluble volatile residue is less than 5000 ppm, refer to Remark #2.

CENTRIFUGATION METHOD

This method is applicable to samples containing suspended matter which are very difficult to filter.

1) Apparatus Centrifugal separator about 2000 rpm. Precipitation tube 50 to 100 ml.

2) Operation Pour a proper amount of test water (so as to make suspended matter exceed 5 mg) into the precipitation tube. After balancing each tube, centrifuge at about 2000 rpm for 20 minutes and precipitate the suspended matter in the test water. Remove the supernatant liquid by decanting[3]. To the precipitate add about 10 ml of the

---

[3] When the determination of soluble evaporated residue is performed successively, keep the supernatant liquid. There should be a certain degree of difference in density between the dispersed phase and the dispersion medium to make the application of the centrifuge possible. When a particle of mass mg is centrifuged at an angular velocity of w rad/sec, at a position of r cm from the center of rotation, centrifugal force which the particle receives is as follows. Suppose that the mass of the dispersion medium expelled by a partical is m'g

then $f = (m-m') w^2 r$

Suppose that the specific centrifugal force is RCF and rotational frequency per minute is N (rpm) the water and once again centrifuge and discard the supernatant liquid by decanting.

Transfer the precipitate into an evaporating dish which has been heated to a temperature of 105 to 110°C for 2 hours, allow to cool in a desiccator and weigh. (When chemical balance is used, evaporating dish of the same shape shall be used as supplemental weight after the blank test for it is performed). Then obtain the difference in weight before and after this operation. Calculate the ppm of the suspended matter by the following formula:

$$S = a \times \frac{1000}{V}$$

Where S = Suspended test matter (ppm)
a = Difference in weight before and after evaporation of the test water (mg)
V = Amount of test water (ml)

METHOD OF CALCULATION OF SUSPENDED MATTER FROM THE DIFFERENCE IN EVAPORATED RESIDUE

Calculate the suspended matter from the difference between the total evaporated residue and the soluble evaporated residue.

$$A = B - C$$

Where A = Suspended matters (ppm)
B = Total evaporated residue (ppm)
C = Soluble evaporated residue (ppm)

(3. cont.)
then $$RCF = \frac{F}{(m-m')g} = \frac{W^2r}{g} = 0.00001118\ rN^2$$

From the above equation, it is clear that the centrifugal force near the surface and that at the bottom portion of the liquid are different. For instance, when N = 2000 rpm and the distance between the surface of the liquid in the precipitation tube and the center of rotation is 5 cm (r = 5 cm), RCF is 223g and when the distance between the bottom of precipitation tube and the central axis of rotation is 13 cm, RCF becomes 581g.

Therefore, the RCF value near the surface and that at the bottom shall be reported respectively.

$$\text{Depth of liquid layer } \frac{(\text{RCF at the bottom})\ (\text{RCF at the surface})}{(\text{RCF at the bottom})}$$

x (Distance down to bottom)

In this test, centrifugal separator of which the bottom is 13 cm away from the central axis of rotation at a rotational frequency of 2000 rpm is regarded as the standard.